Massachusetts Institute of Technology

X-ray probes of ordered electronic phases in strongly correlated systems

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Massachusetts Institute of Technology

MIT-BNL workshop, 26 Jul 2017

Outline

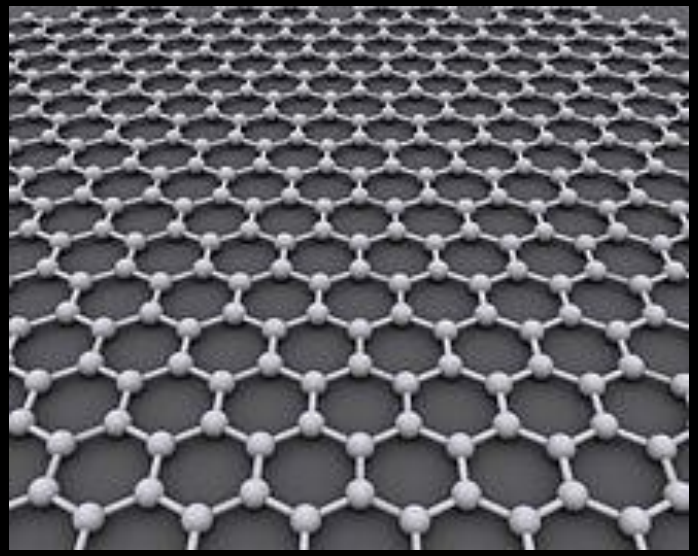
- What is a quantum solids
- Different forms of electronic crystals
- Resonant X-ray Scattering: crystallography for electrons
- Charge order in high-temperature superconductors
- The frontier: Coherent X-ray nanoscattering and imaging

What *is* a quantum solid?

Defining a quantum solid is not easy

Graphite? *No*

Graphene? **Yes**



What *is* a quantum solid?

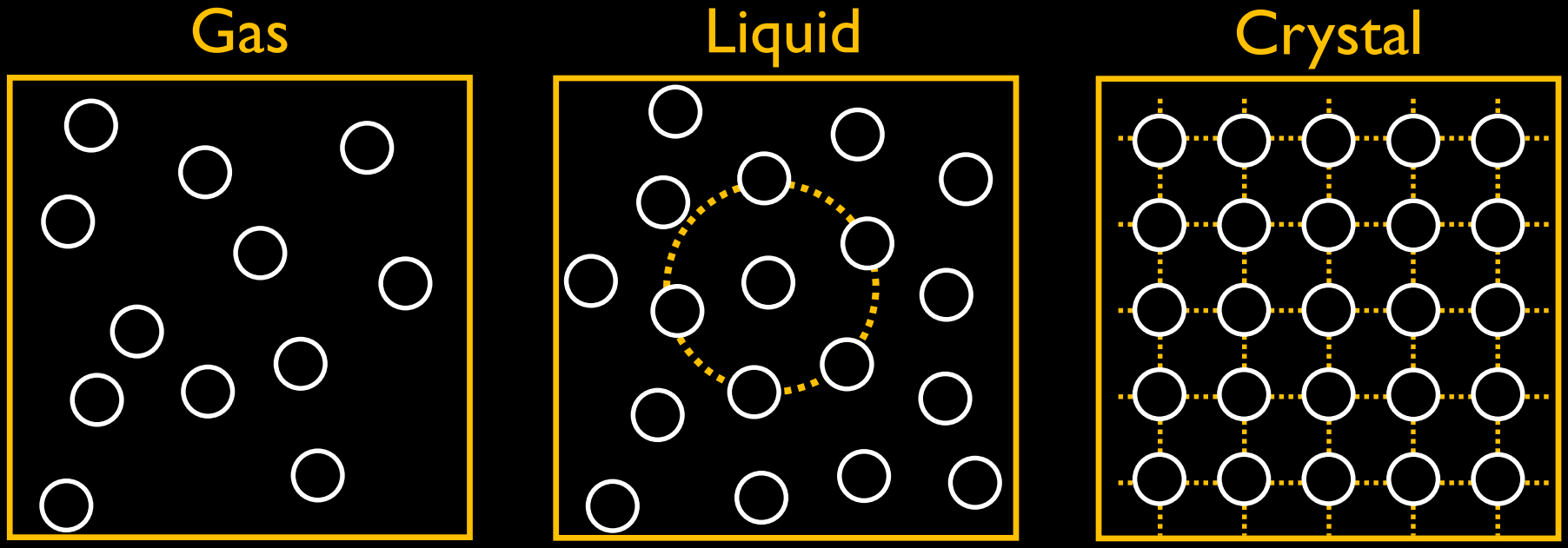
Defining a quantum solid is not easy

> “I shall not today attempt further to define the kinds of material I understand to be embraced within that shorthand description [“*quantum solid*"], and perhaps I could never succeed in intelligibly doing so. But *I know it when I see it* ”

Improbably readapted from Justice Stewart, *378 U.S. at 197*

Strongly interacting systems

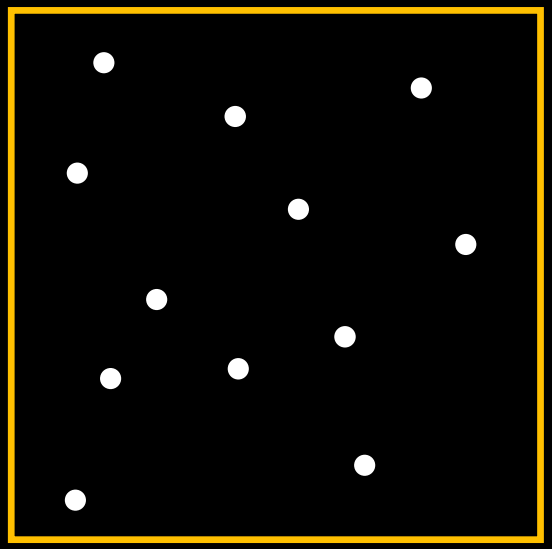
$U \ll kT$ | Kinetic energy (T) ⟷ Interaction energy (U) | $U \gg kT$



Strongly interacting systems

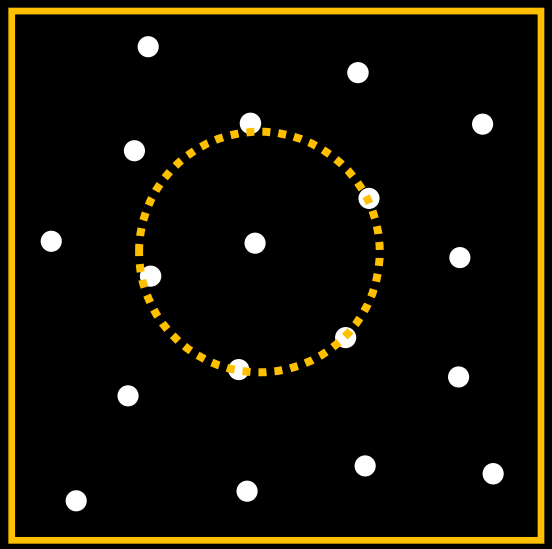
$U \ll kT$ | Kinetic energy (T) ⟷ Interaction energy (U) | $U \gg kT$

Electron Gas



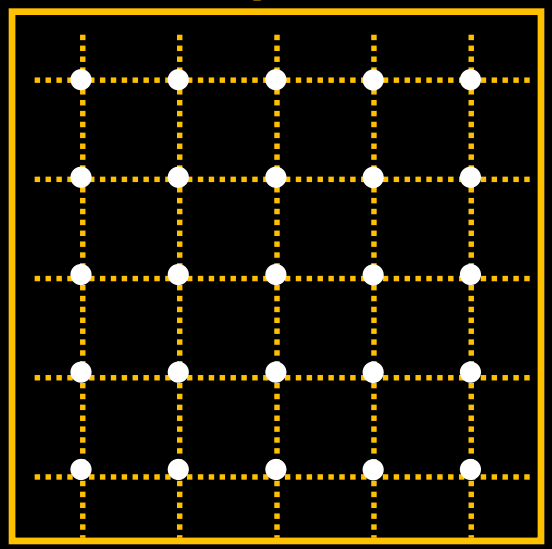
Disorder

Electron Liquid



Short-range order

Electron Crystal



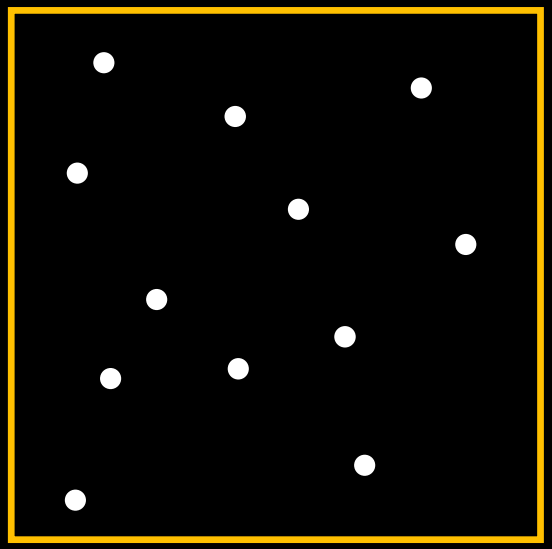
Long-range order

Order from disorder

Strongly interacting systems

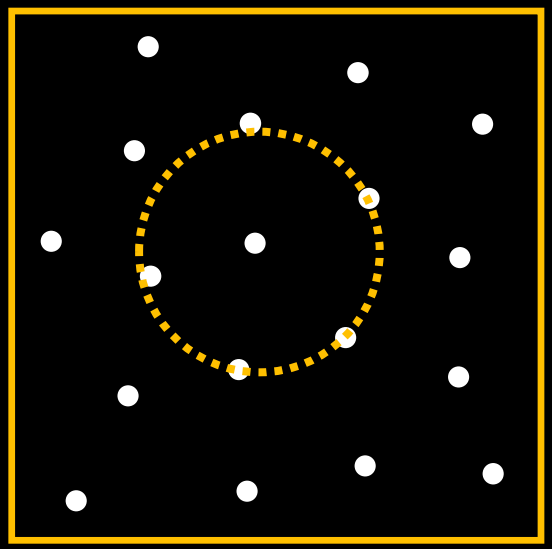
$U \ll kT$ | Kinetic energy (T) ⟷ Interaction energy (U) | $U \gg kT$

Electron Gas



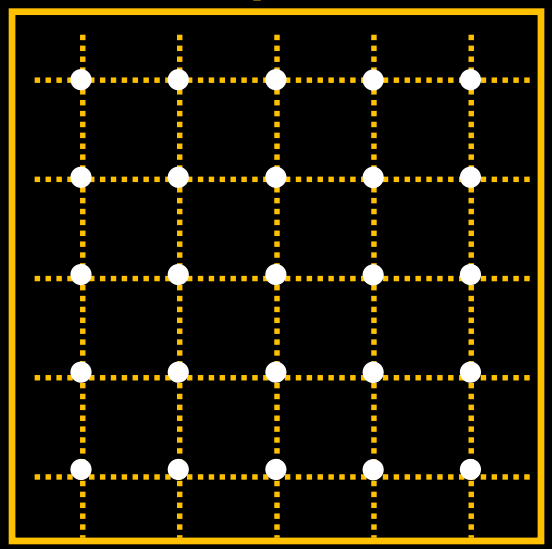
Disorder

Electron Liquid



Short-range order

Electron Crystal



Long-range order

Order from disorder

Strongly interacting systems

$U \ll kT$

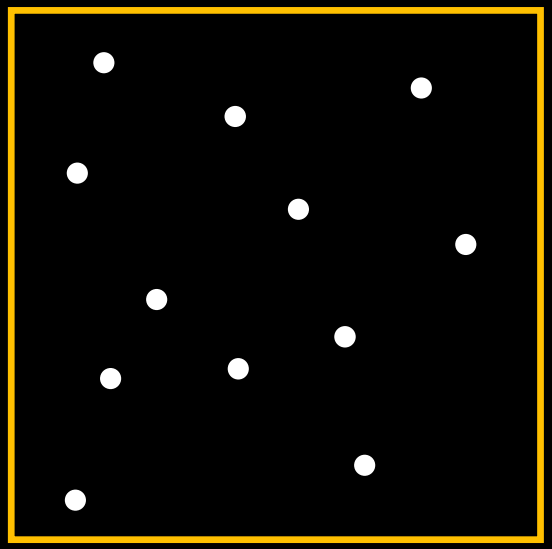
Kinetic energy (T) ⟷ Interaction energy (U)

Cold atoms: nK

2DEG: mK

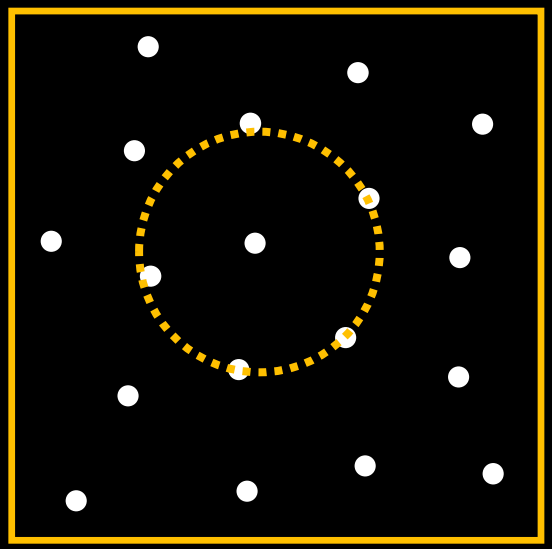
SCES: 10-10^3 K

Electron Gas



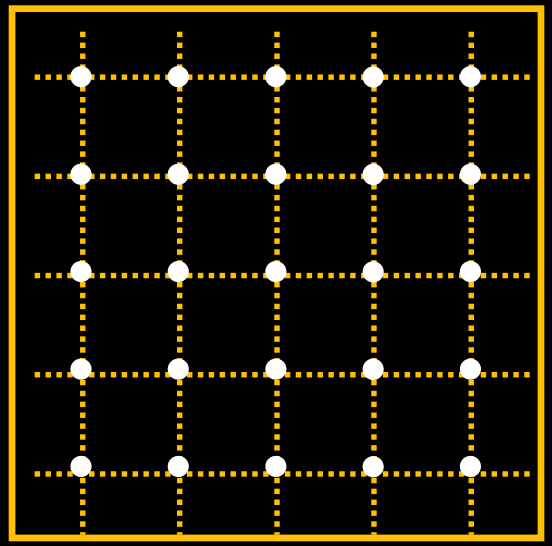
Disorder

Electron Liquid



Short-range order

Electron Crystal

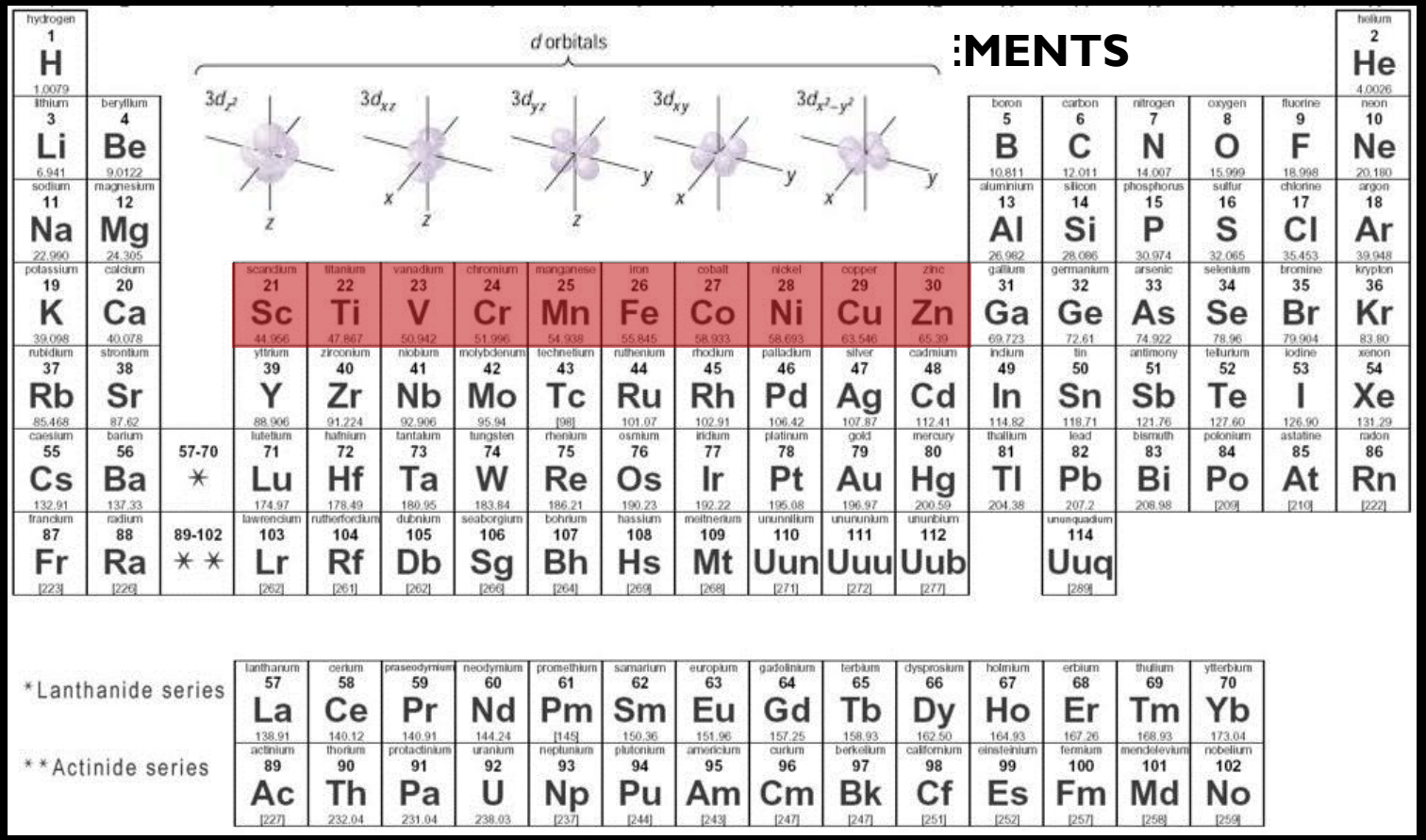


Long-range order

Order from disorder

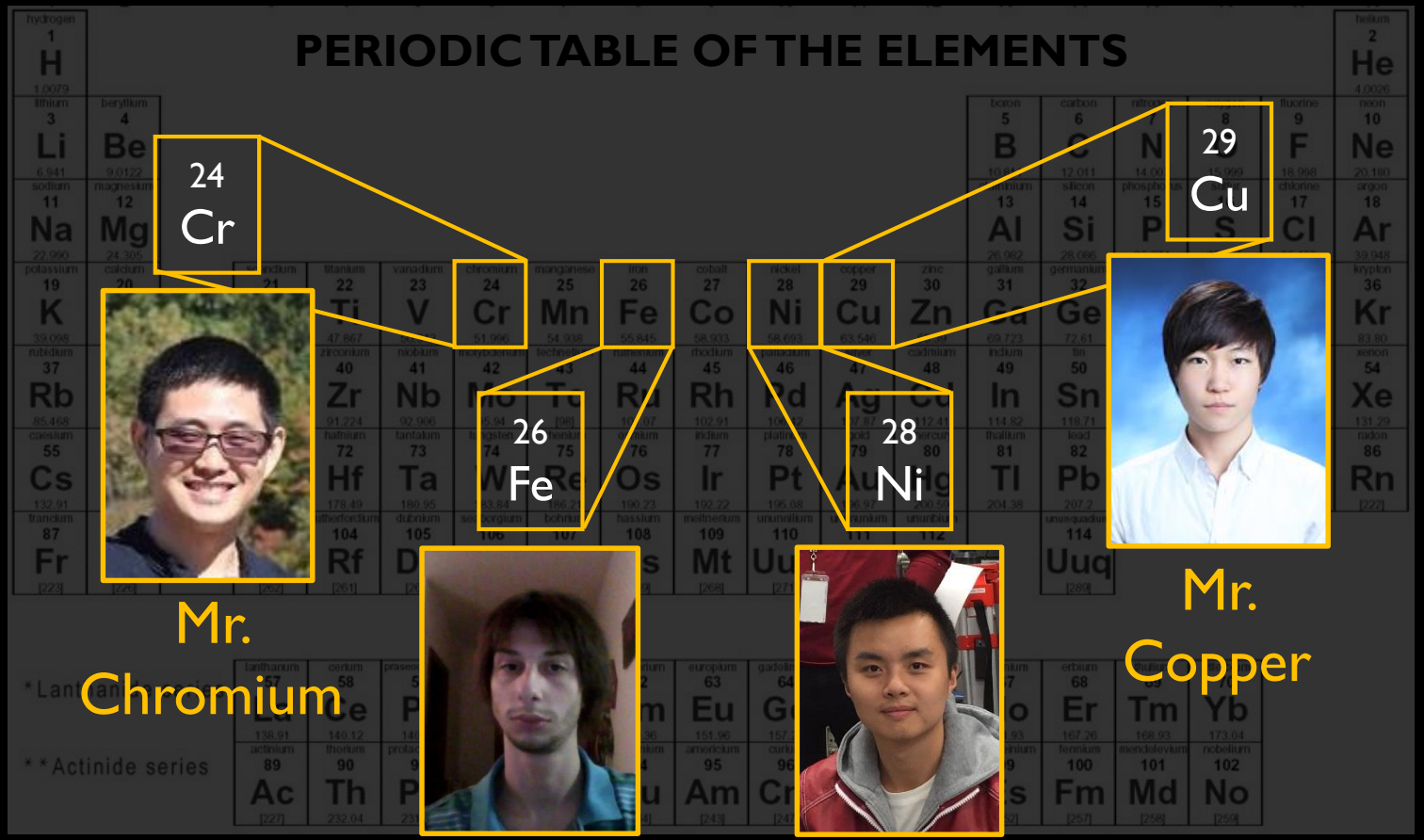
Strongly interacting systems

Strong interactions (U) ⟺ Localized orbitals



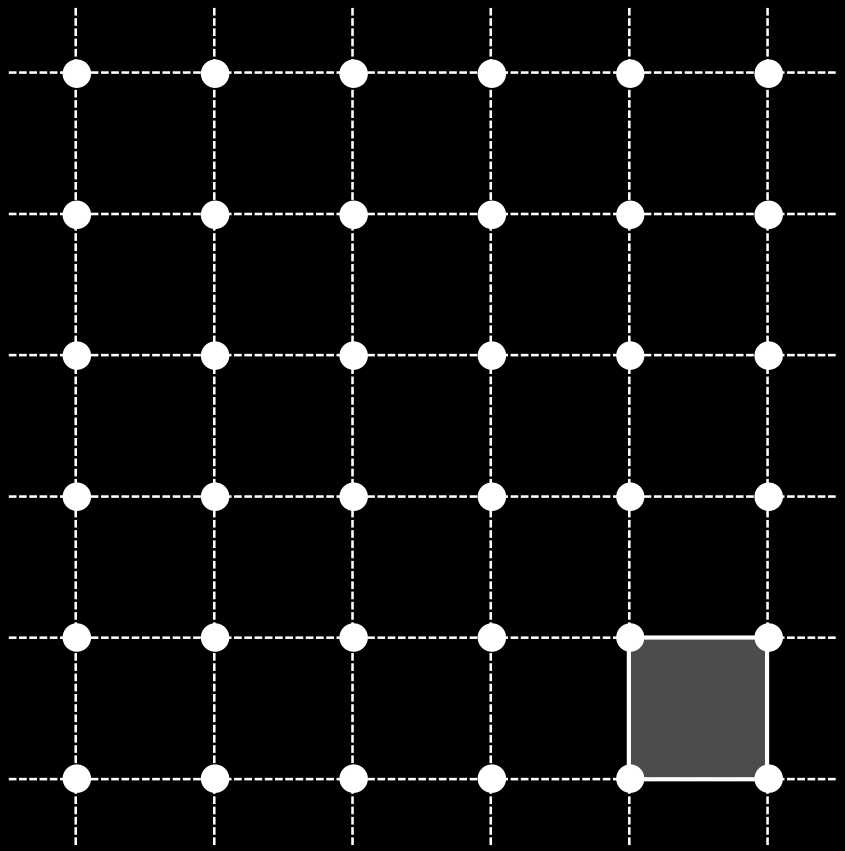
Strongly interacting systems

Strong interactions (*U*) ⟷ Localized orbitals



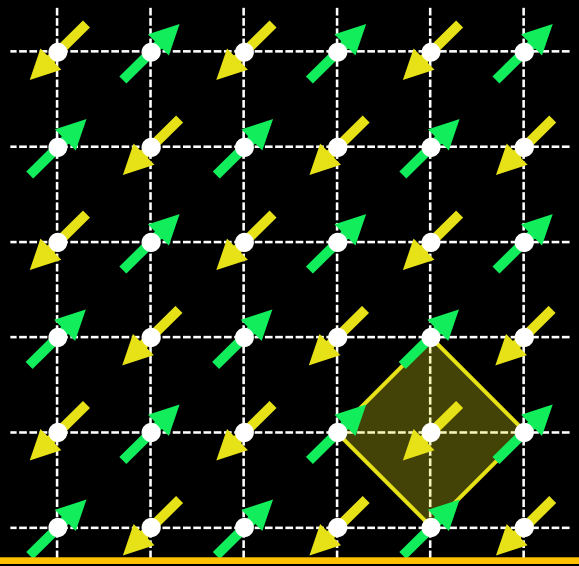
Ordered states of matter

Crystalline

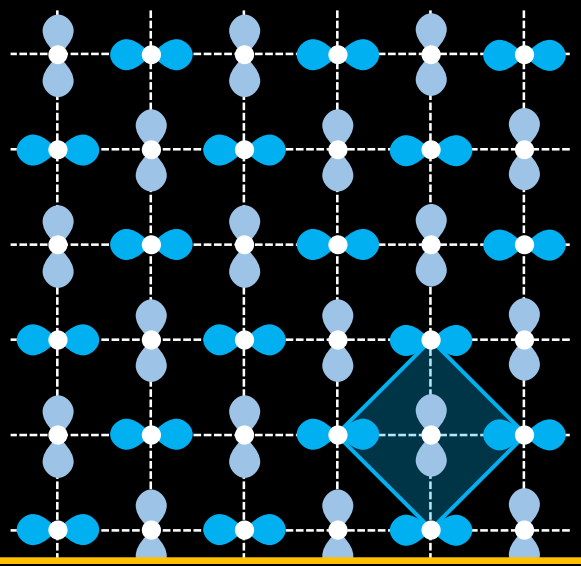


Symmetries defined by space group

(Anti-ferro)magnetic

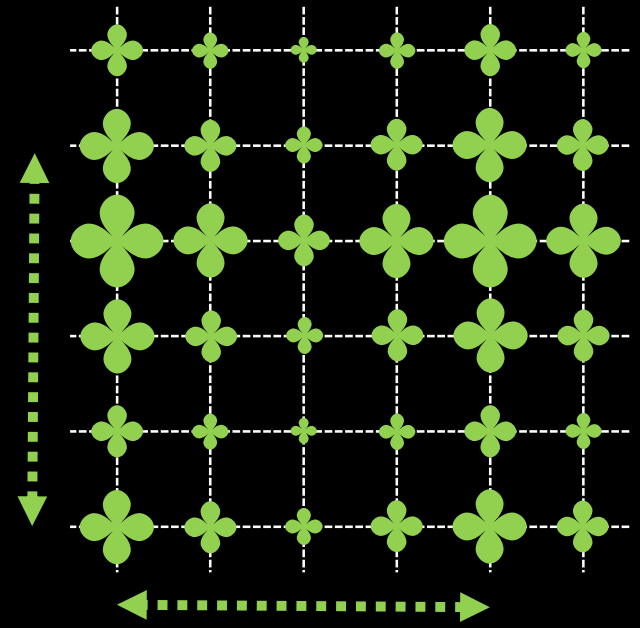


Orbital

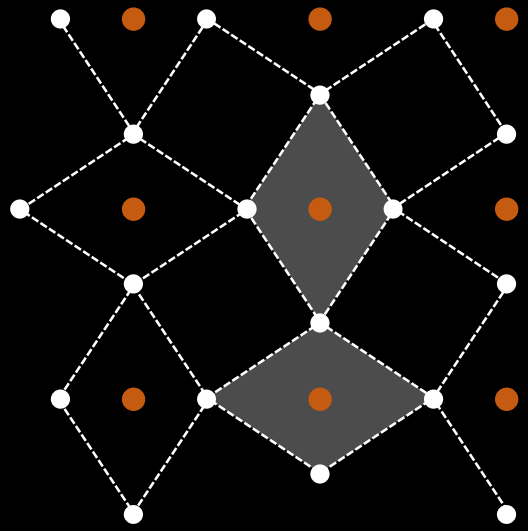


Native symmetries of atomic lattice are broken!

Charge



Lattice

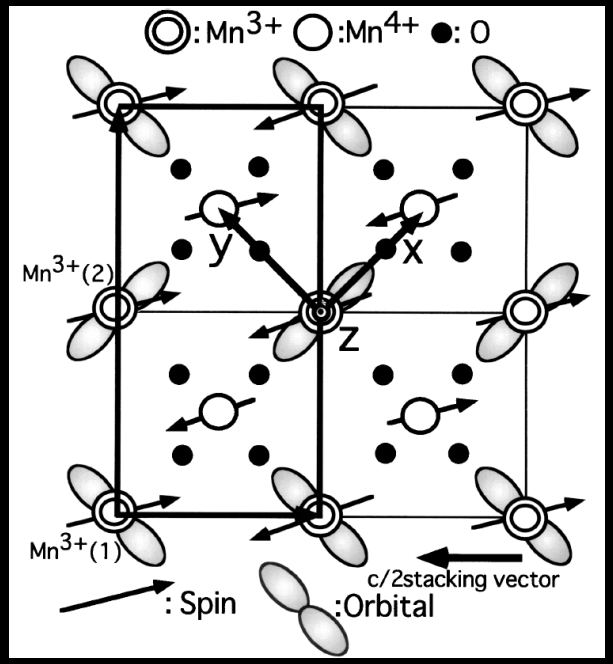


Electronic crystals

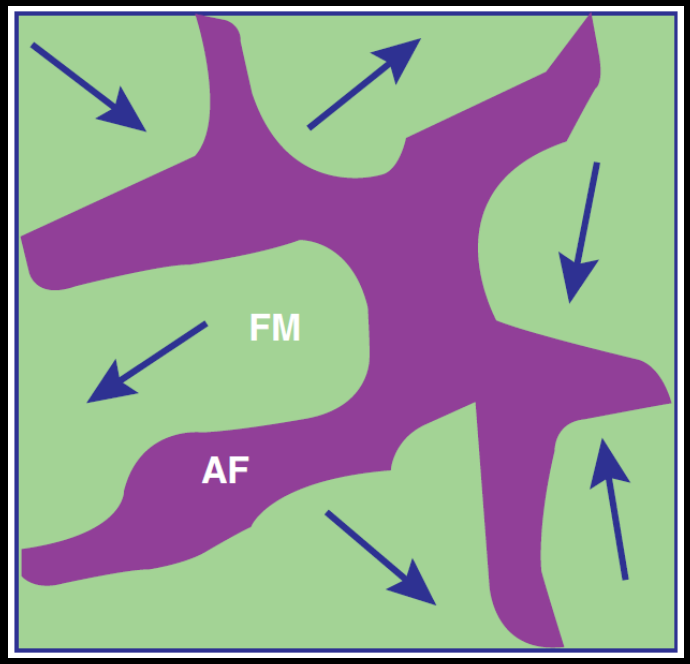
Phase coexistence *vs* Phase separation

$La_{0.5}Sr_{1.5}MnO_4$
spin/charge/orbital
orders all at once!

FM and AFM orders
competing to become
ground state

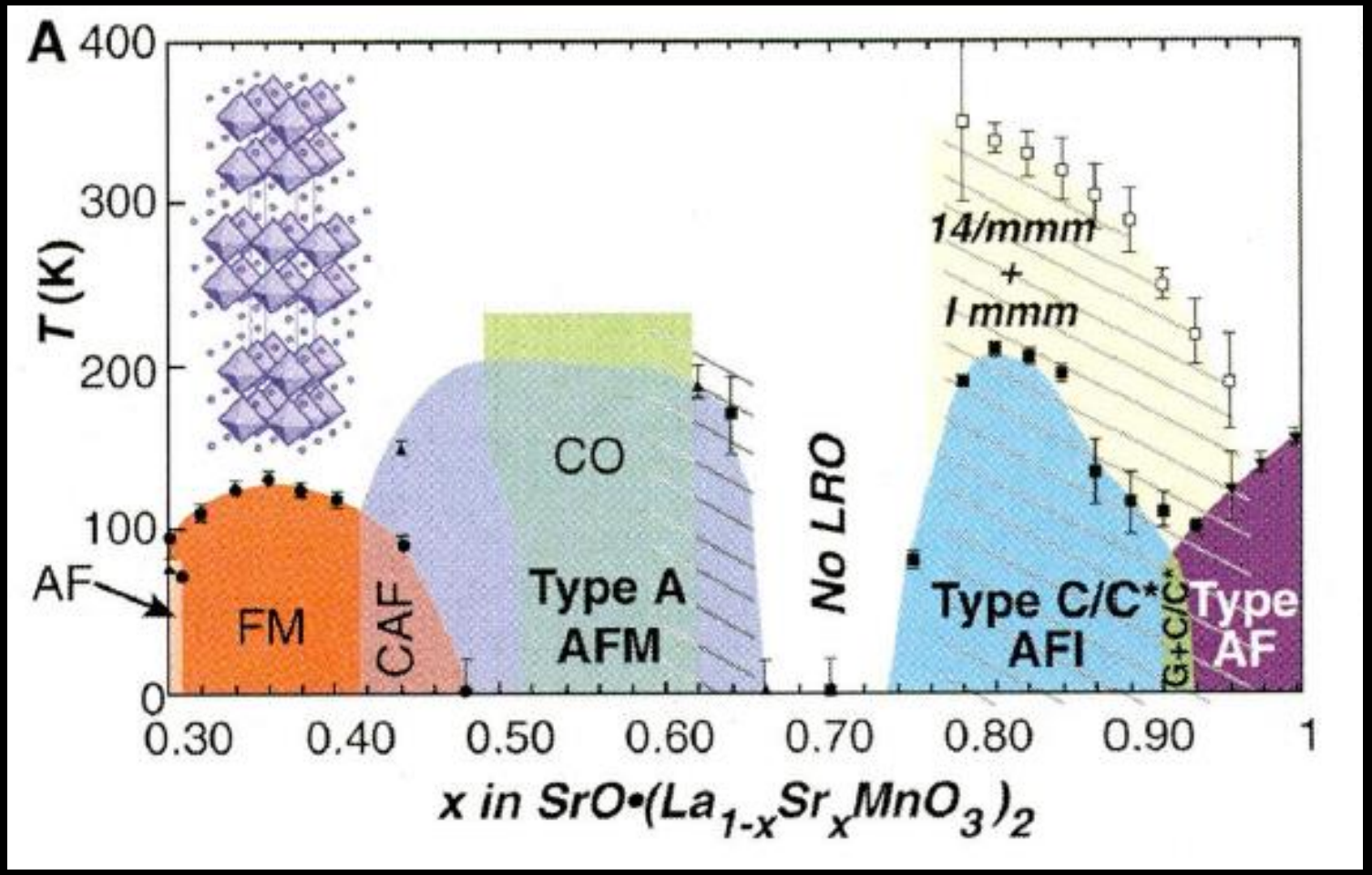


Murakami *et al.*, *PRL* **80**, 1932 (1998)



E. Dagotto, *Science* **309**, 257 (2005)

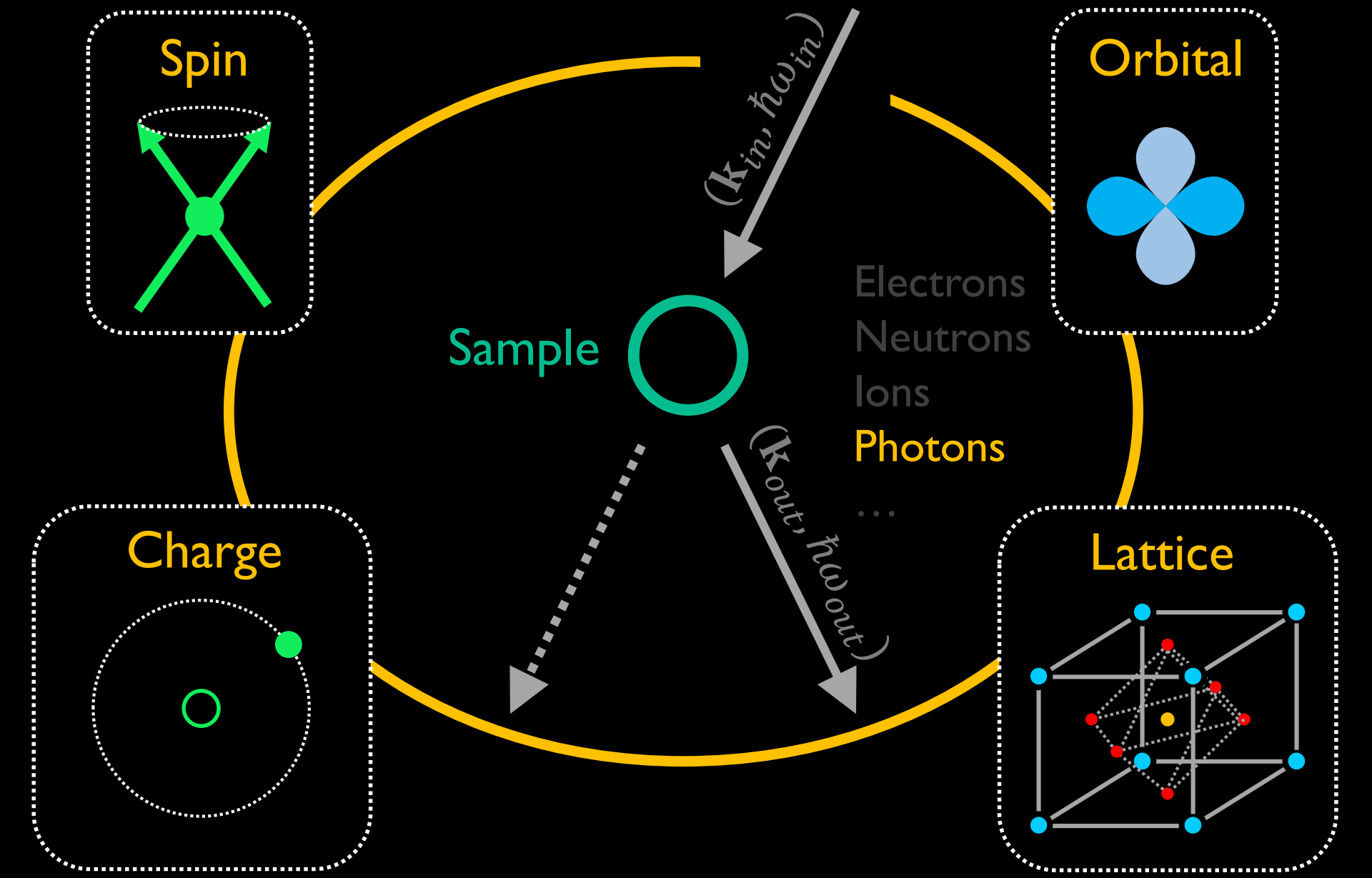
Electronic crystals



E. Dagotto, *Science* **309**, 257 (2005)

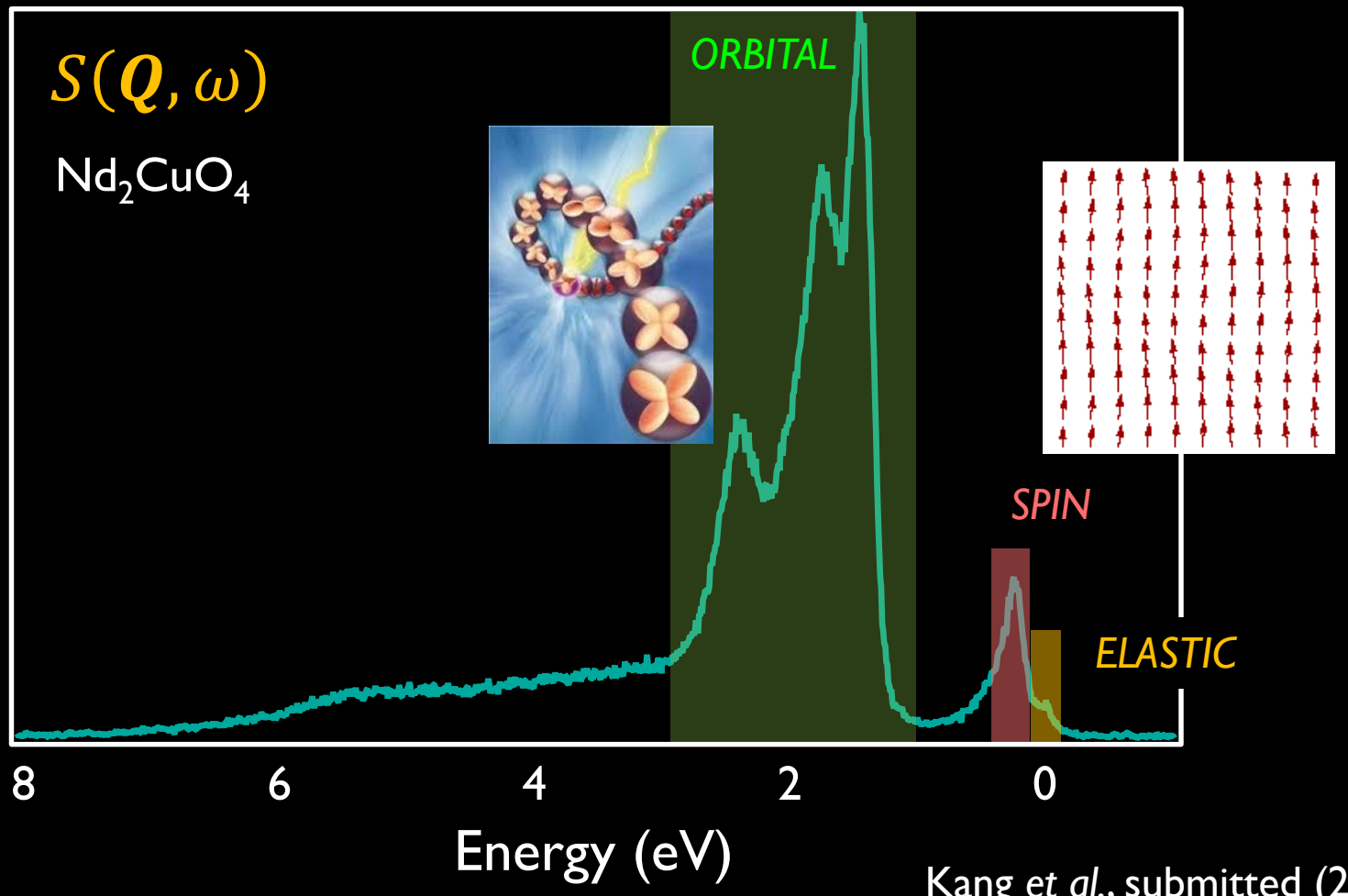
How do we detect electronic crystals?

Scattering probes



Scattering probes

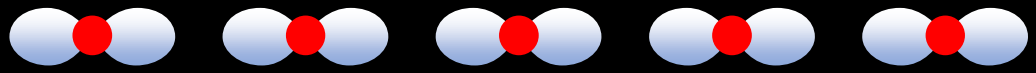
We can detect static orders ($\omega = 0$) but also dynamical excitations ($\omega \neq 0$)



Kang *et al.*, submitted (2017)

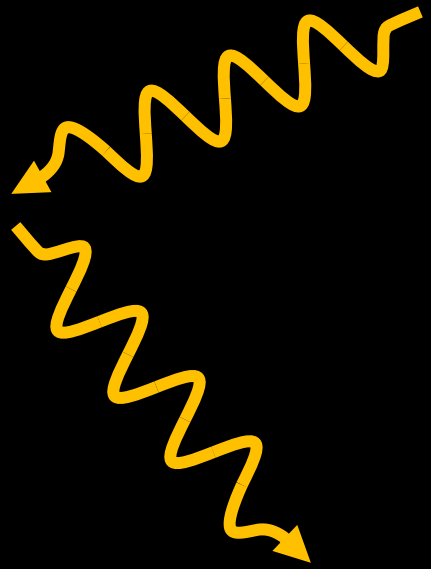
Scattering probes

Valence electrons (shallow)

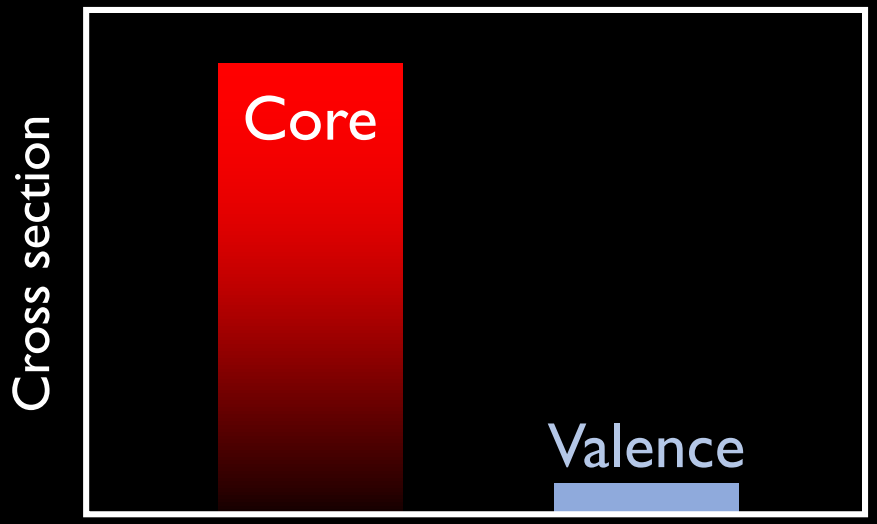


Core electrons (deep, tightly bound)

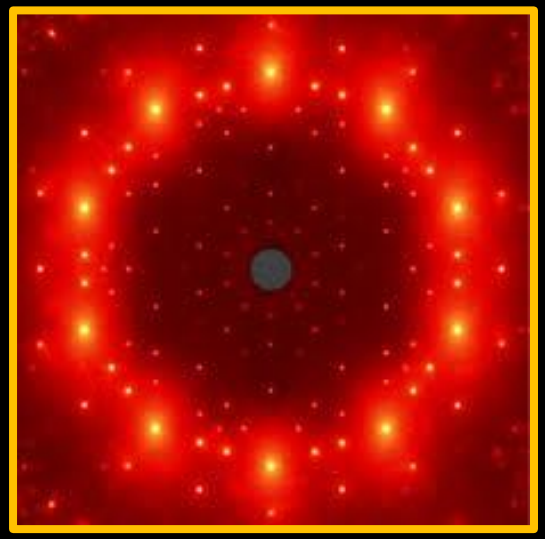
Lattice position →



Cross section ~ Z (no. of electrons)



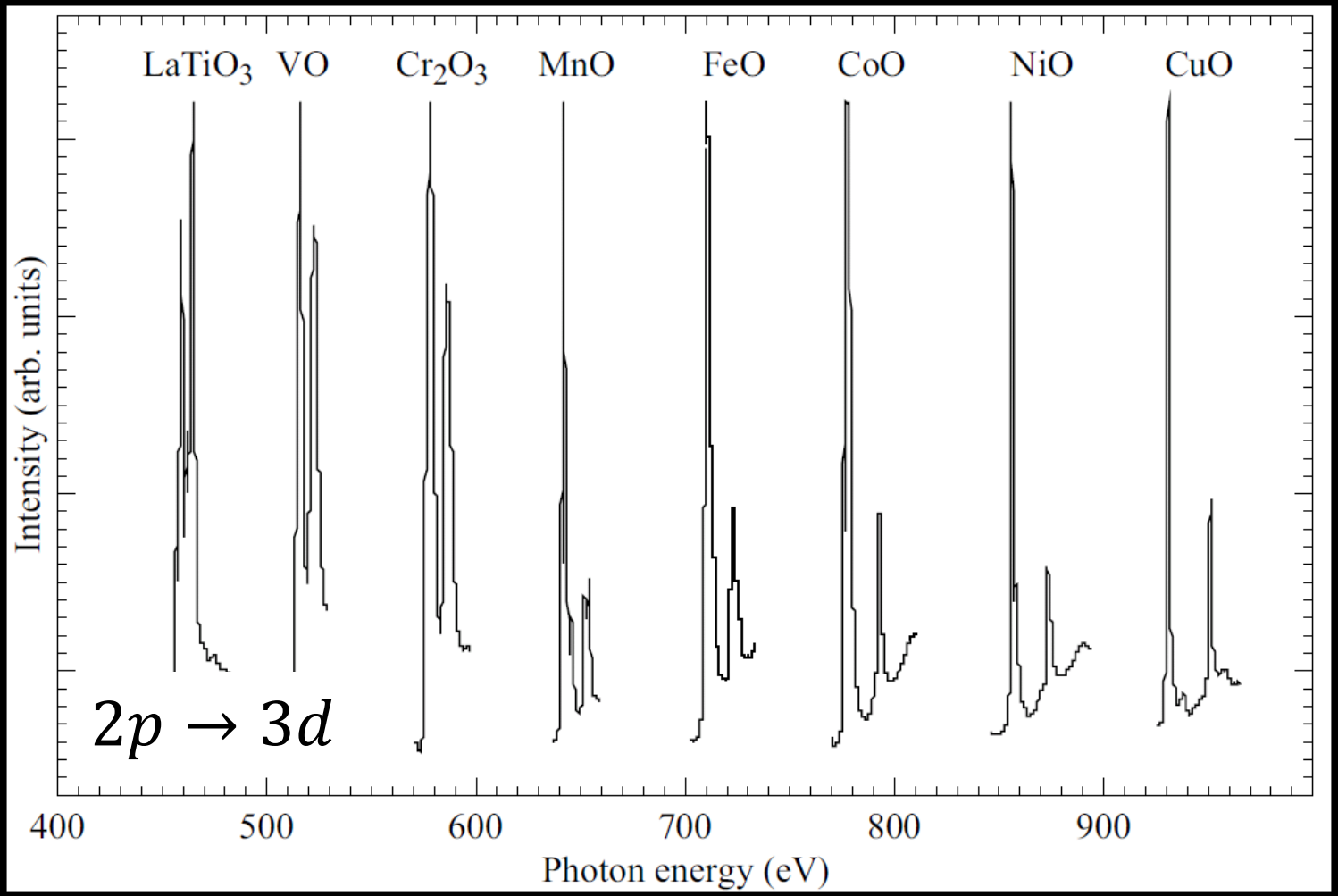
Core electrons dominate XRD signal



XRD – X-Ray Diffraction

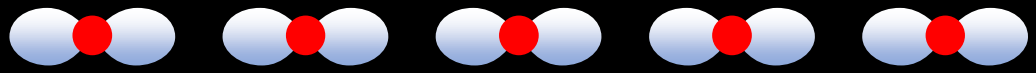
Scattering probes

22	23	24	25	26	27	28	29
Ti	V	Cr	Mn	Fe	Co	Ni	Cu



Resonant scattering

Valence electrons (shallow)



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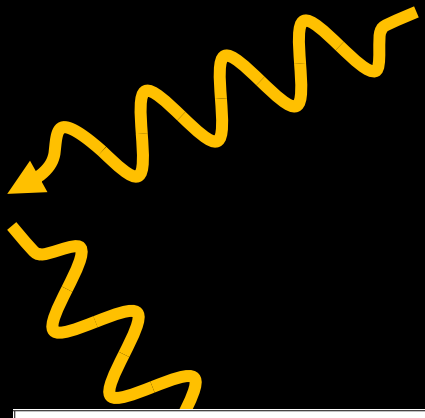
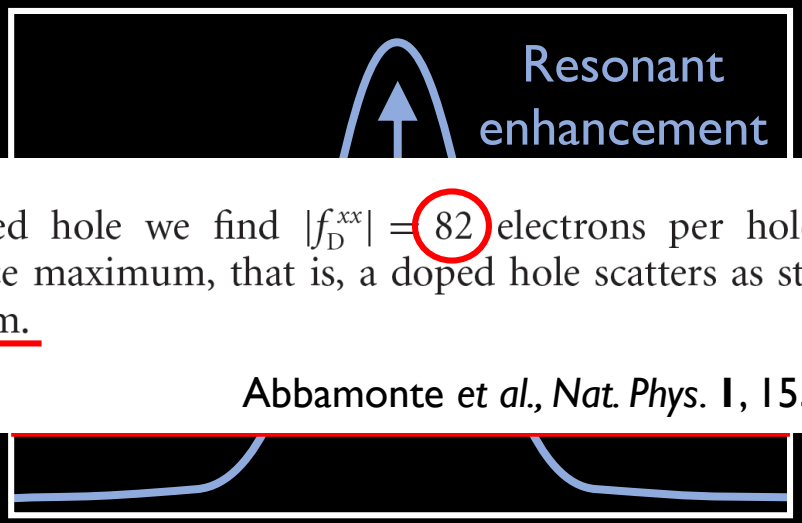


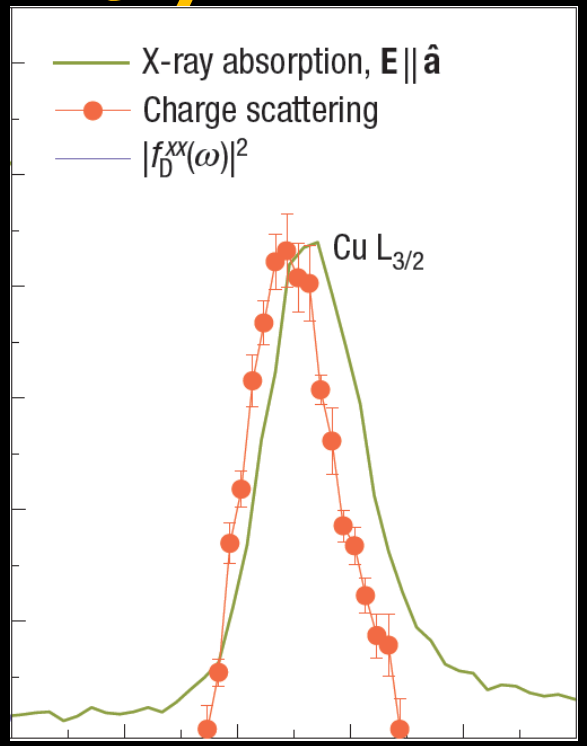
Lattice position →



For an *x*-oriented hole we find $|f_D^{xx}| = 82$ electrons per hole on the resonance maximum, that is, a doped hole scatters as strongly as a Pb atom.

Abbamonte *et al.*, *Nat. Phys.* **1**, 155 (2005)

Photon energy ($h\nu$)



Photon energy ($h\nu$)

Scattering probes

There is no better place than NSLS-II

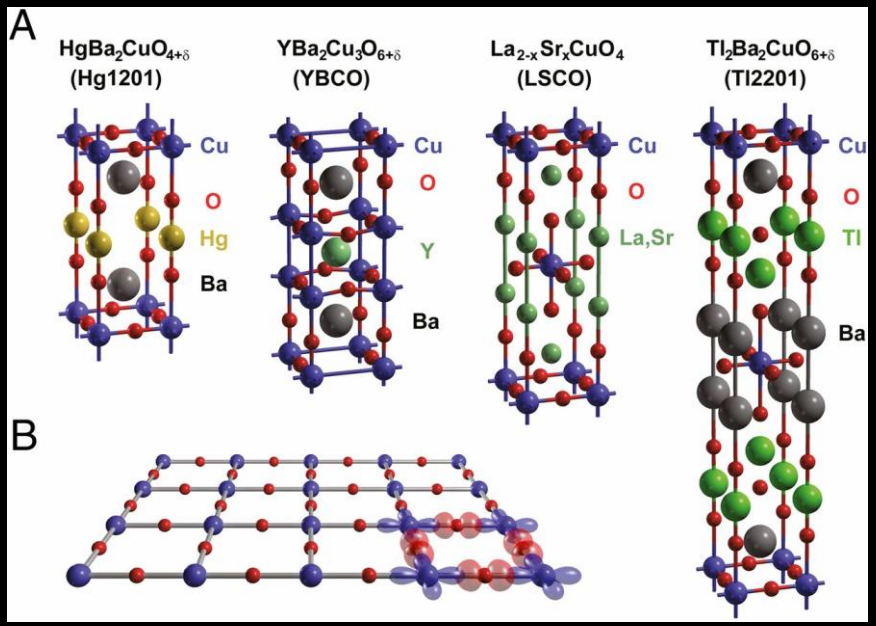
- Beamline CSX-1:
  - World's brightest and fully coherent facility for soft x-ray scattering beamline (~10^{13} ph/s).

- Beamline SIX:
  - Resonant inelastic x-ray scattering with highest energy resolution (60000 resolving power)

Electronic orders in copper oxide high-temperature superconductors (*cuprates*)

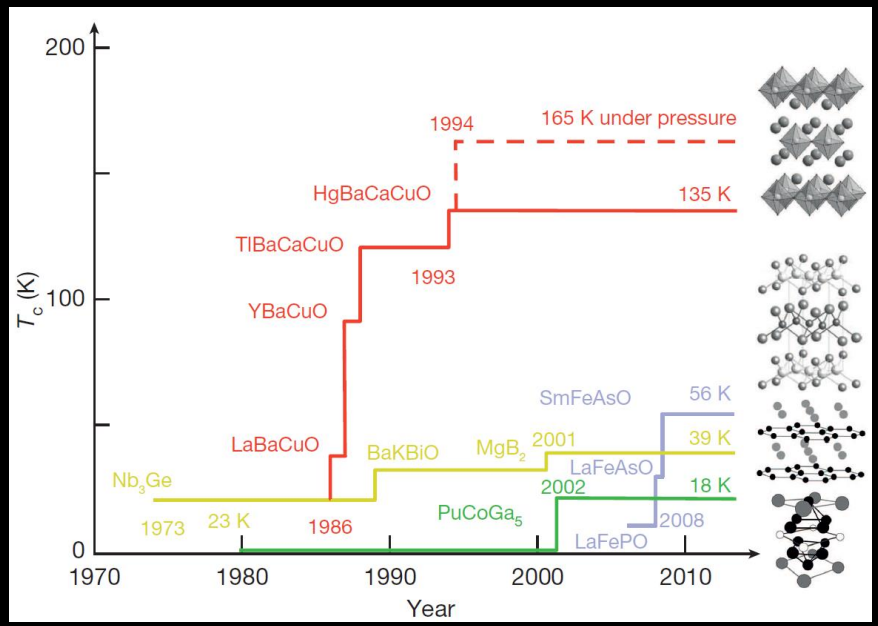
Cuprates: a favourite physicist's playground

Structure



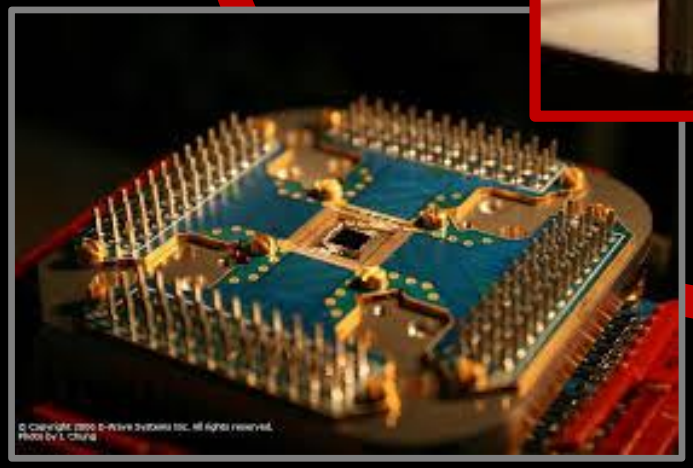
Barisic *et al.*, *PNAS* **110,** 12235 (2013)

Superconductivity

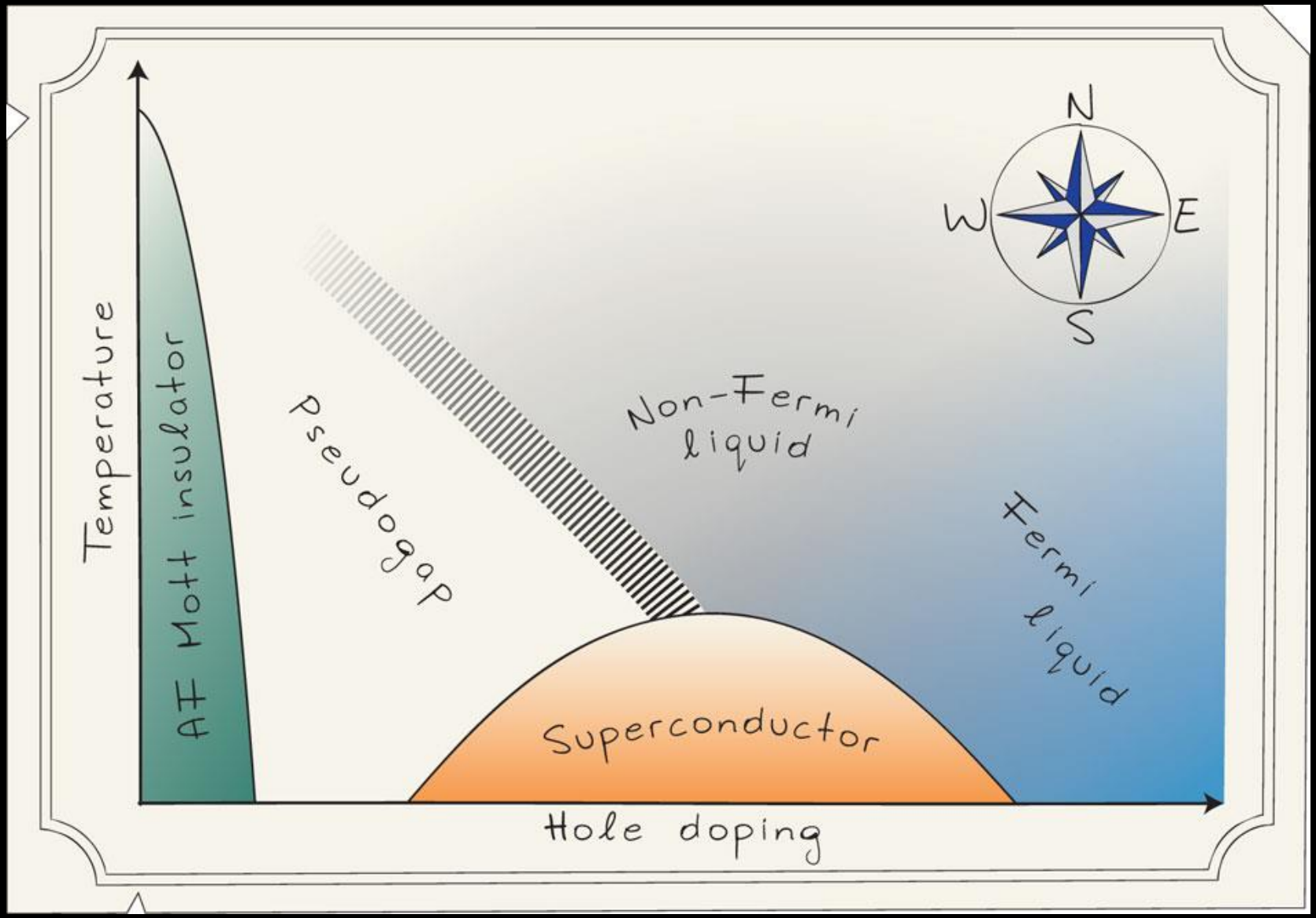


Keimer *et al.*, *Nature* **518,** 179 (2015)

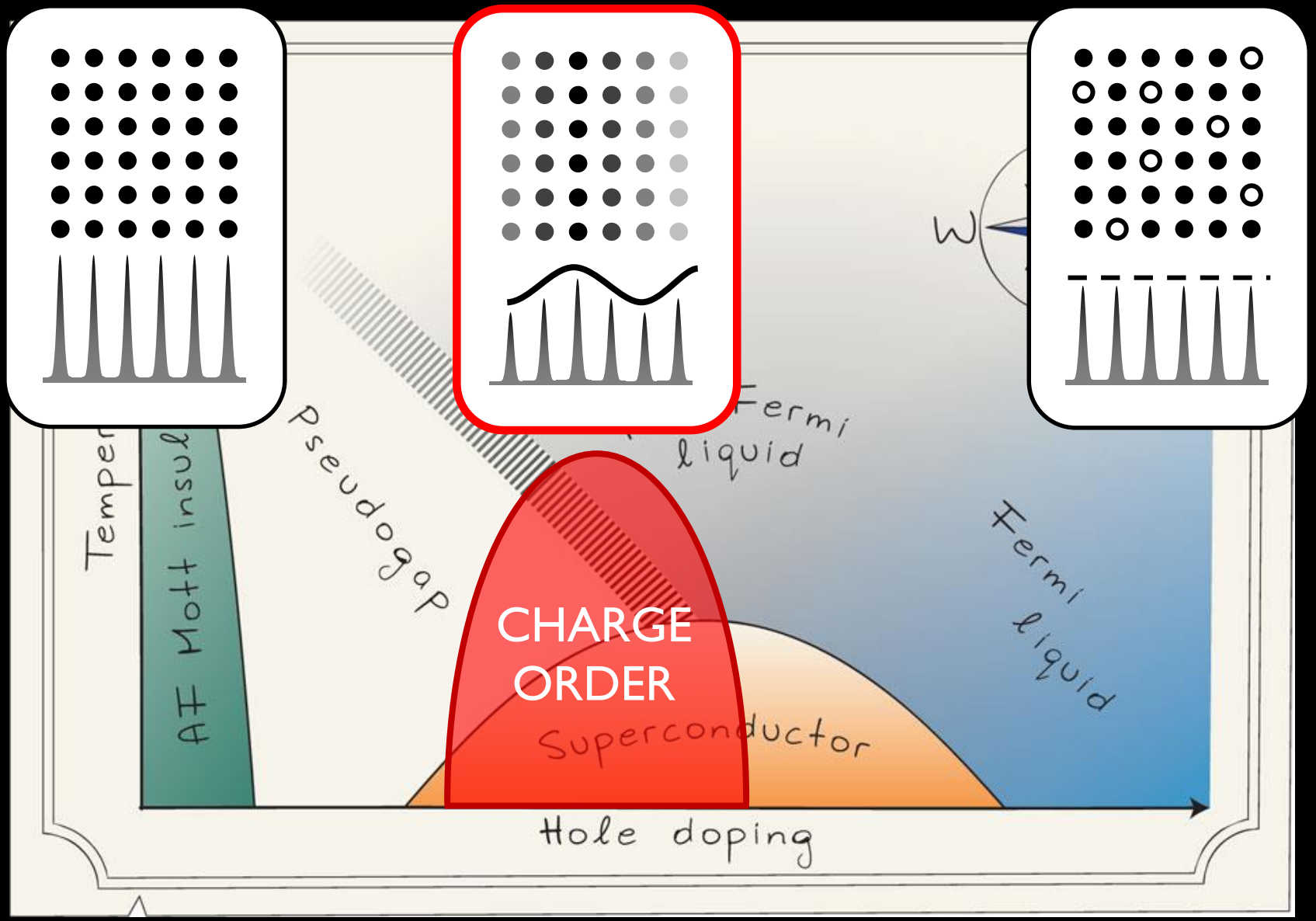
Cuprates: a favourite physicist's playground



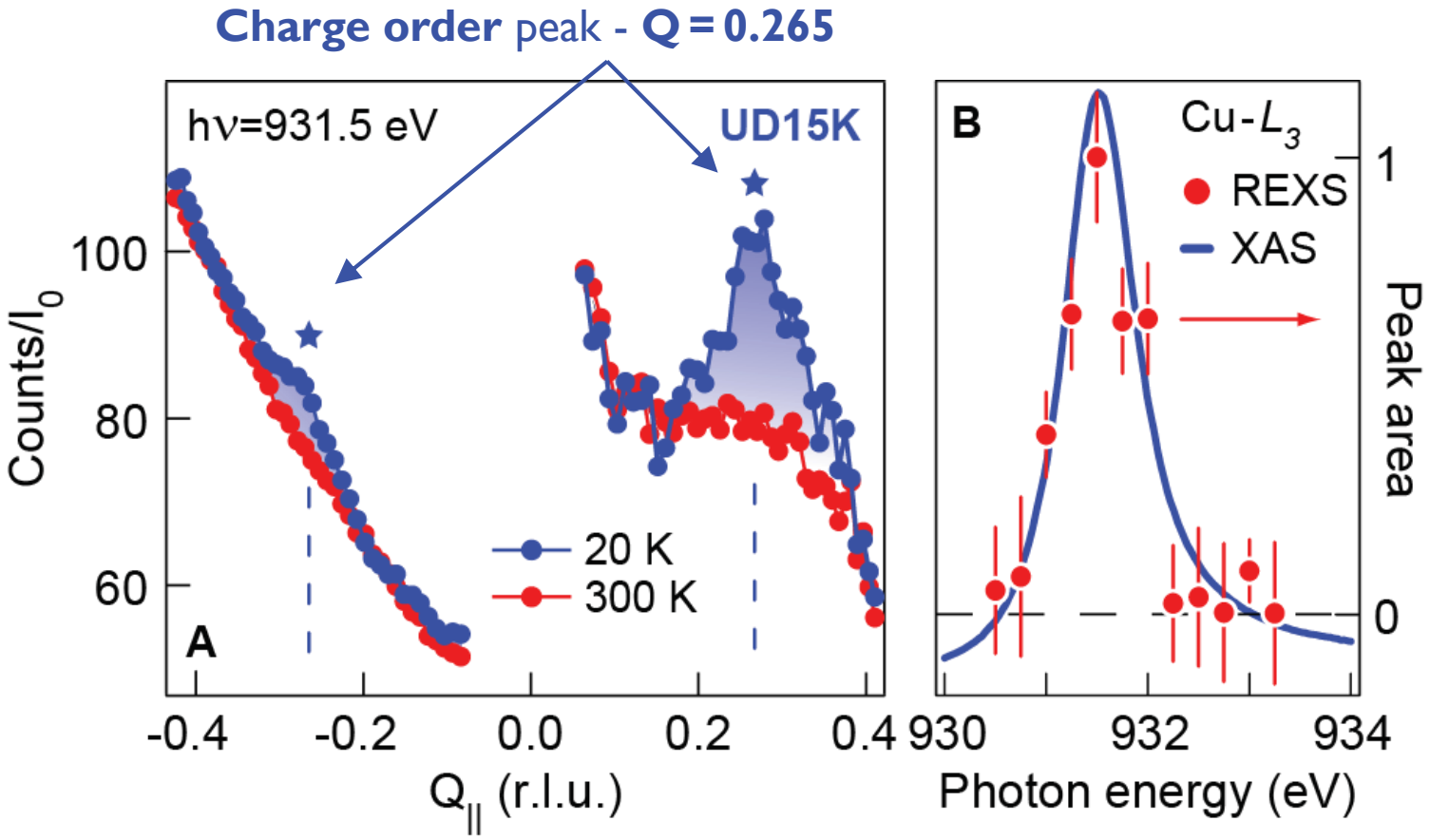
Cuprates: a favourite physicist's playground



Cuprates: a favourite physicist's playground



Charge order in cuprates

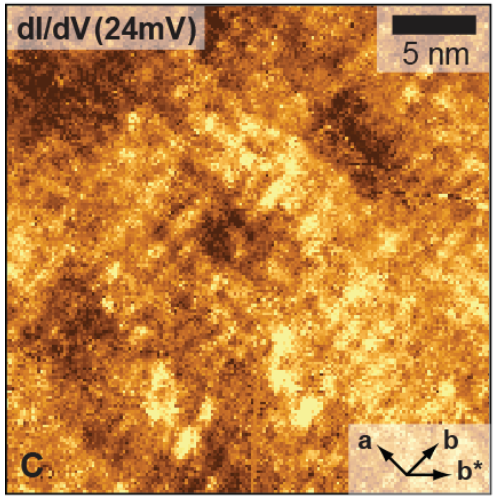


Working near resonance is key!

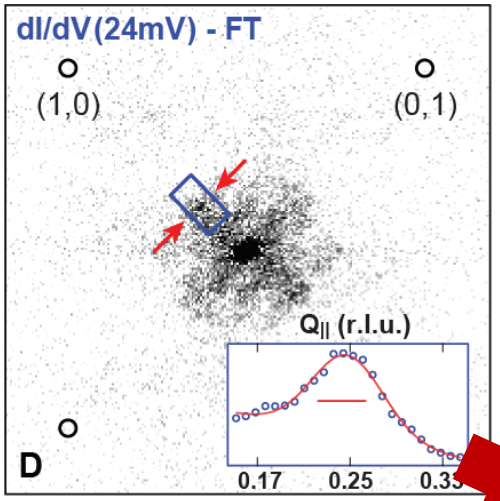
Comin *et al*, *Science* **340**, 390-392 (2014)

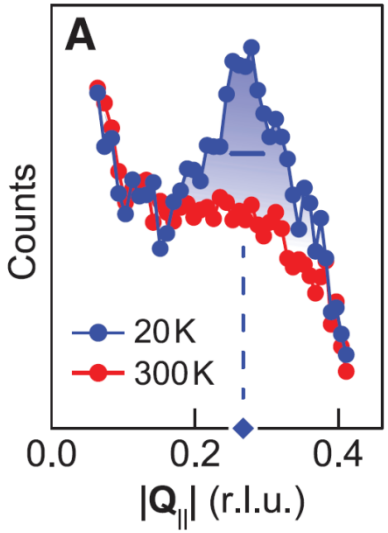
Charge order in cuprates

Real space

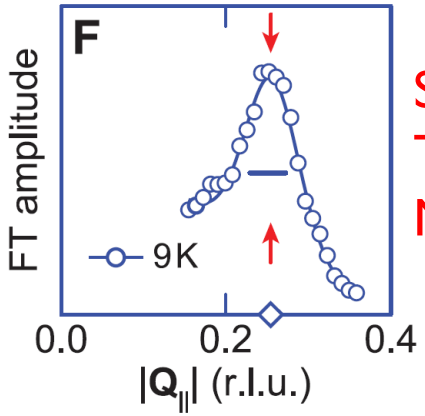


Fourier transform





Resonant X-ray Scattering



Scanning Tunneling Microscopy

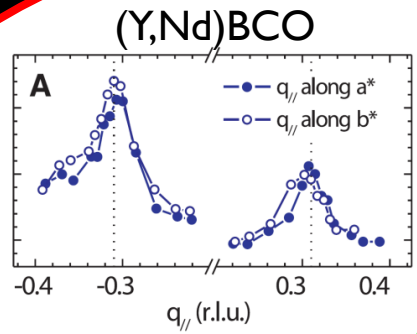
Jenny Hoffman's lab

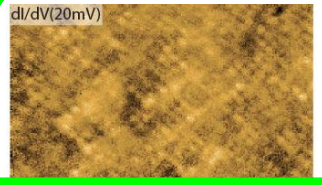
Comin *et al*, *Science* **340**, 390-392 (2014)

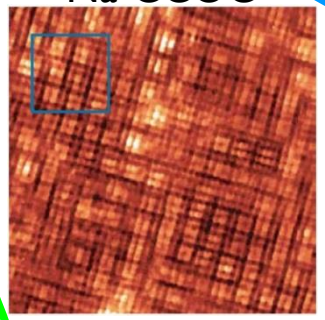
Charge ordering in Bi2201 – RXS and STM

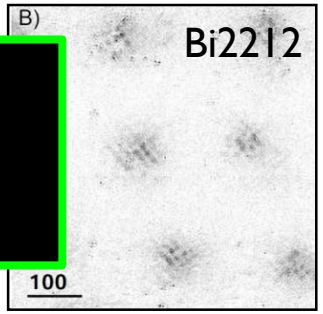
SCATTERING
BULK

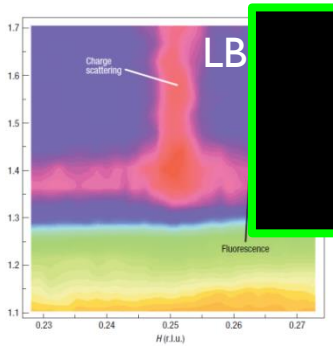
STM
SURFACE

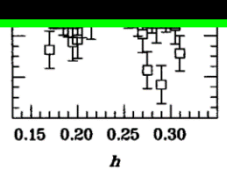


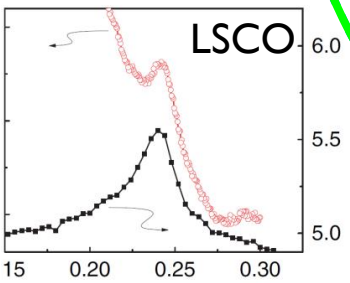


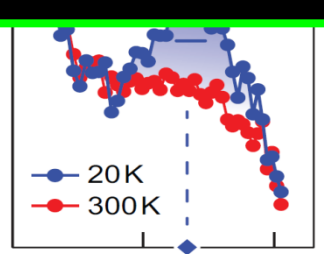


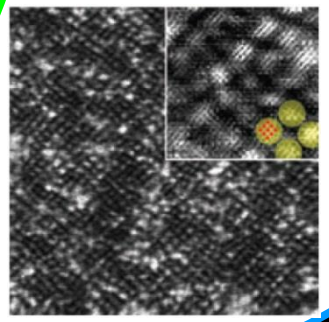












Charge order is a universal phenomenon in the cuprates

Why *resonant* scattering?

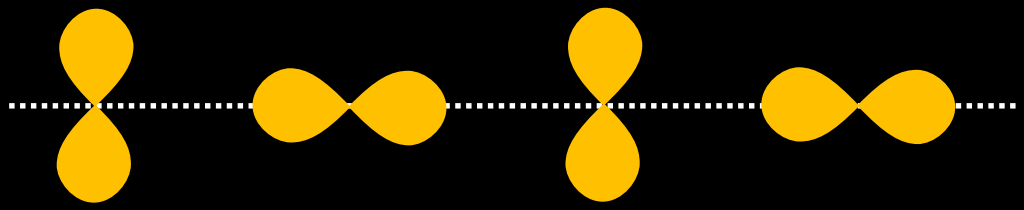
The inner symmetry of charge order

$$I_{\mathrm{XRD}}(\boldsymbol{Q}) \propto \left|(\varepsilon' \cdot \varepsilon^*) \sum_i f_i(\boldsymbol{Q}) e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_i}\right|^2 \qquad f_i(\boldsymbol{Q}) \xrightarrow[Q\to 0]{} Z_i$$

SCALAR

$$I_{\mathrm{RXS}}(\boldsymbol{Q}) \propto \left|\varepsilon'_\alpha \cdot \left(\sum_i f_i^{\alpha\beta}(\boldsymbol{Q}, E) e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_i}\right) \cdot \varepsilon^*_\beta\right|^2$$

TENSOR



Phase factor	1	-1	1	-1
Charge	Q	Q	Q	Q
Form factor	f_A	f_B	f_A	f_B

$$I_{\mathrm{XRD}} \propto Q \times (1 - 1 + 1 - 1) = 0$$

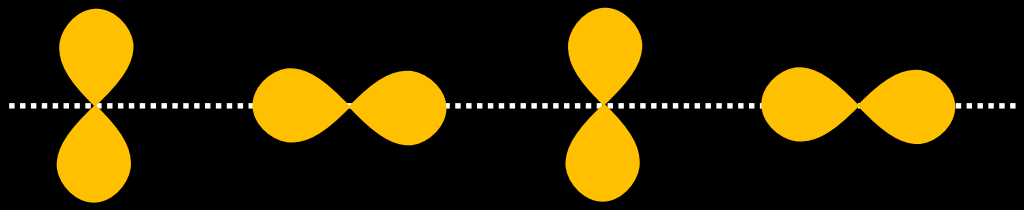
The inner symmetry of charge order

$$I_{\mathrm{XRD}}(\boldsymbol{Q}) \propto \left|(\varepsilon' \cdot \varepsilon^*) \sum_i f_i(\boldsymbol{Q}) e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_i}\right|^2 \qquad f_i(\boldsymbol{Q}) \xrightarrow[Q\to 0]{} Z_i$$

SCALAR

$$I_{\mathrm{RXS}}(\boldsymbol{Q}) \propto \left|\varepsilon'_\alpha \cdot \left(\sum_i f_i^{\alpha\beta}(\boldsymbol{Q}, E) e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_i}\right) \cdot \varepsilon^*_\beta\right|^2$$

TENSOR

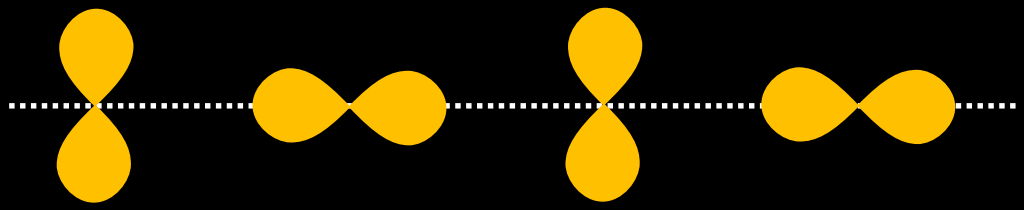


Phase factor	I	-I	I	-I
Charge	Q	Q	Q	Q
Form factor	f_A	f_B	f_A	f_B

$$I_{\mathrm{RXS}} \propto f_A \cdot (1+1) - f_B \cdot (1+1) = 2 \cdot (f_A - f_B) \neq 0$$

The inner symmetry of charge order

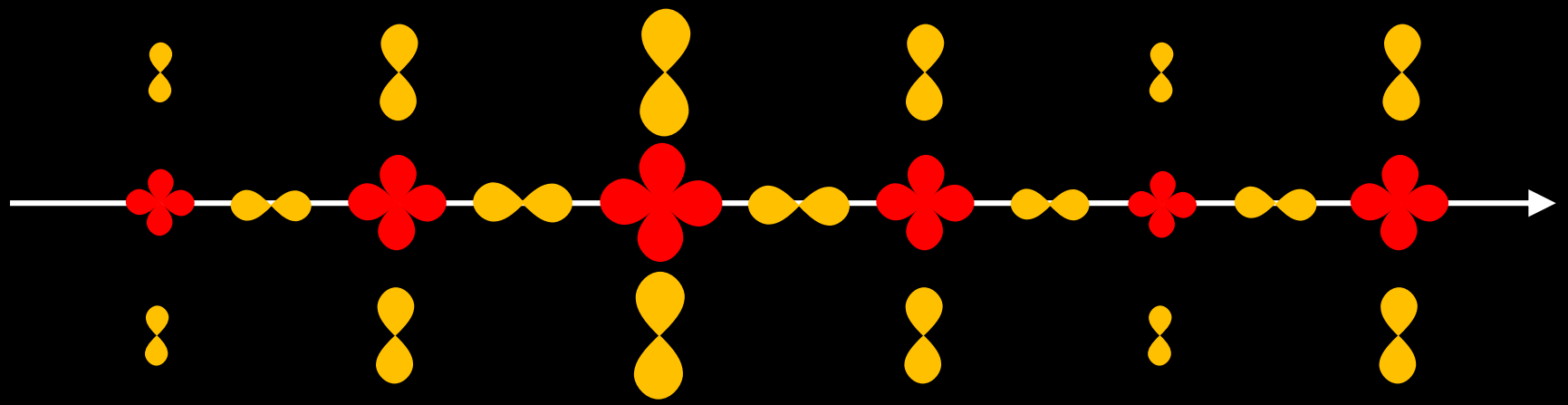
Access the *symmetry* of the charge (or spin/orbital) distribution

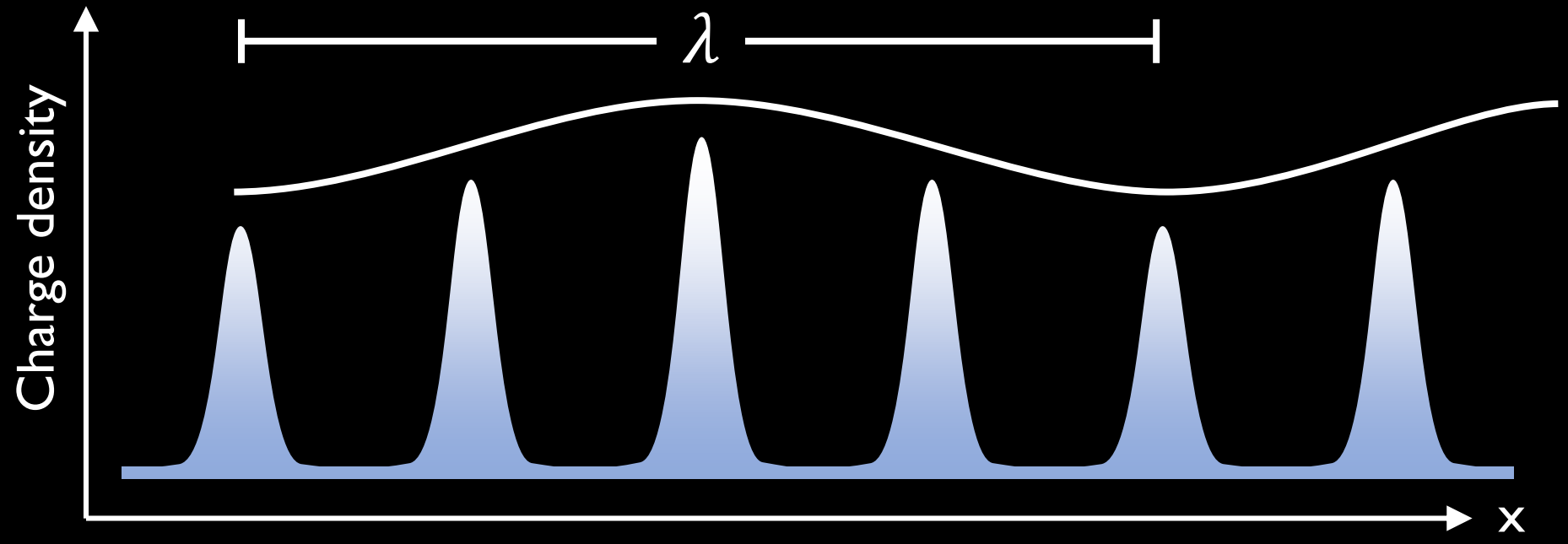


Phase factor	1	-1	1	-1
Charge	Q	Q	Q	Q
Form factor	f_A	f_B	f_A	f_B

$$I_{\mathrm{RXS}} \propto f_A \cdot (1+1) - f_B \cdot (1+1) = 2 \cdot (f_A - f_B) \neq 0$$

The inner symmetry of charge order



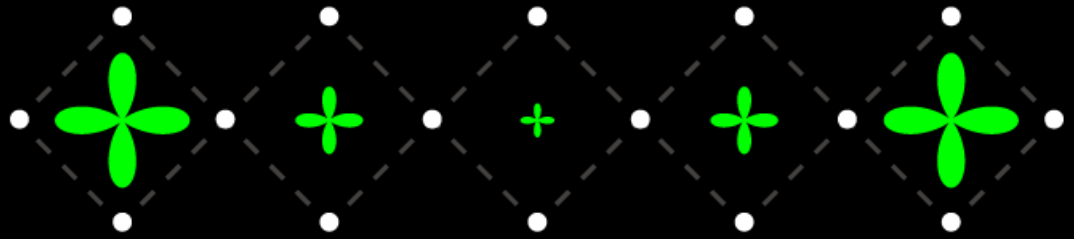


The inner symmetry of charge order

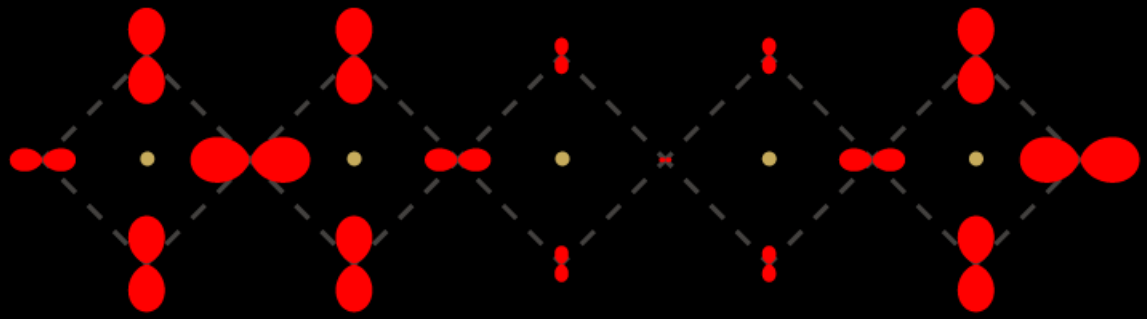
$\boldsymbol{k}-$dependence

$$\Delta_{CDW}(\boldsymbol{k},\boldsymbol{Q}) = \delta(\boldsymbol{Q}-\boldsymbol{Q}^*)\cdot[\Delta_s + \Delta_{s'}(\cos k_x + \cos k_x) + \Delta_d(\cos k_x - \cos k_x)]$$

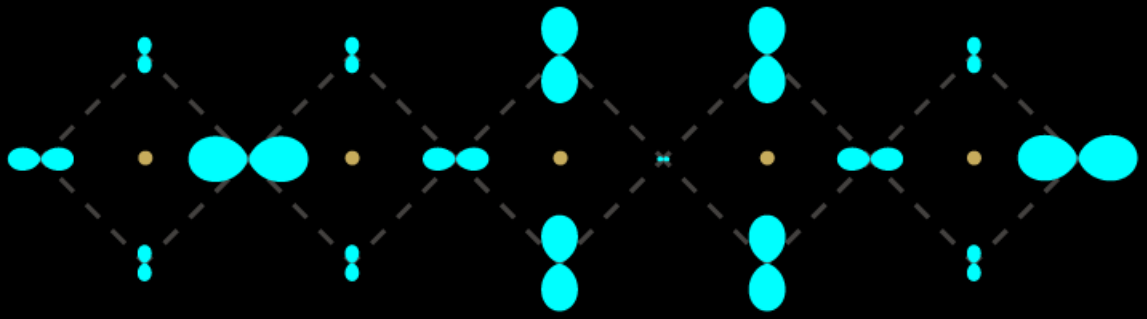
Site (*s*)



Bond (*s'*)



Bond (*d*)



The inner symmetry of charge order

$$F_{pq}(\pm \boldsymbol{Q}_{CO}) =$$

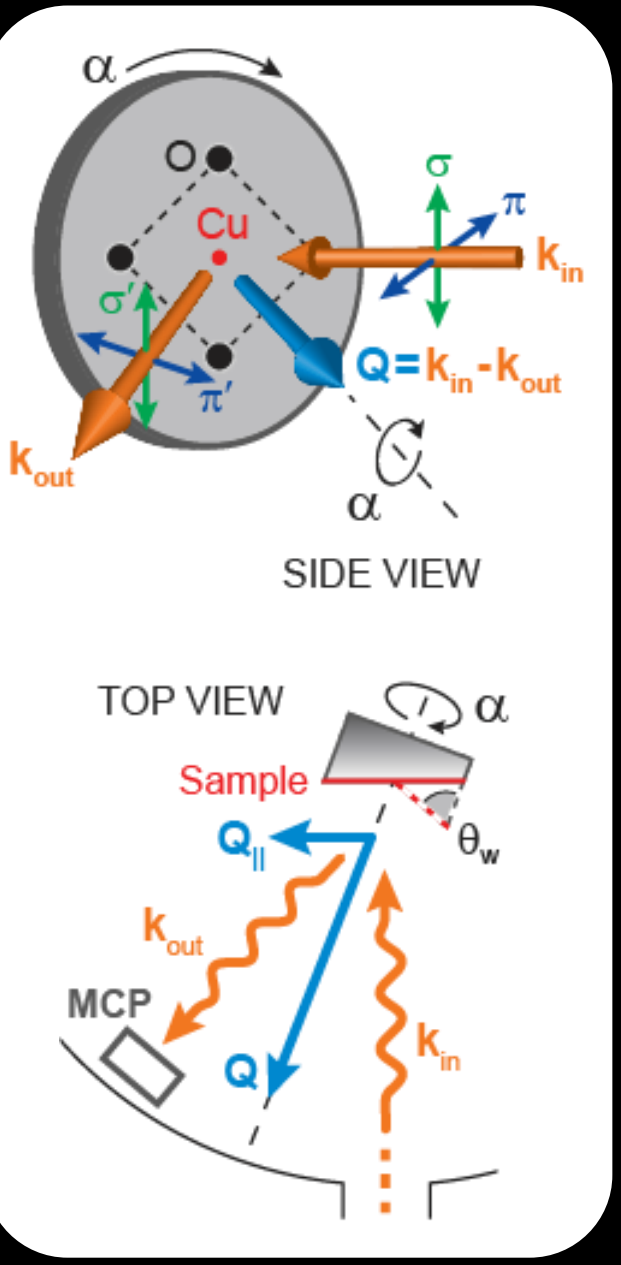
$$\delta_s \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \gamma \end{pmatrix}$$

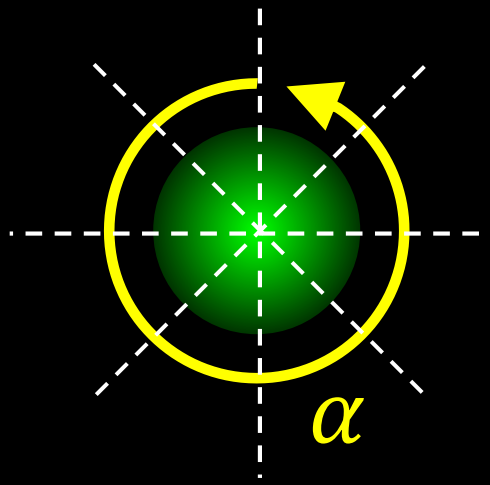
$$\delta_{s'} \begin{pmatrix} \cos\phi & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

$$\delta_d \begin{pmatrix} \cos\phi & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

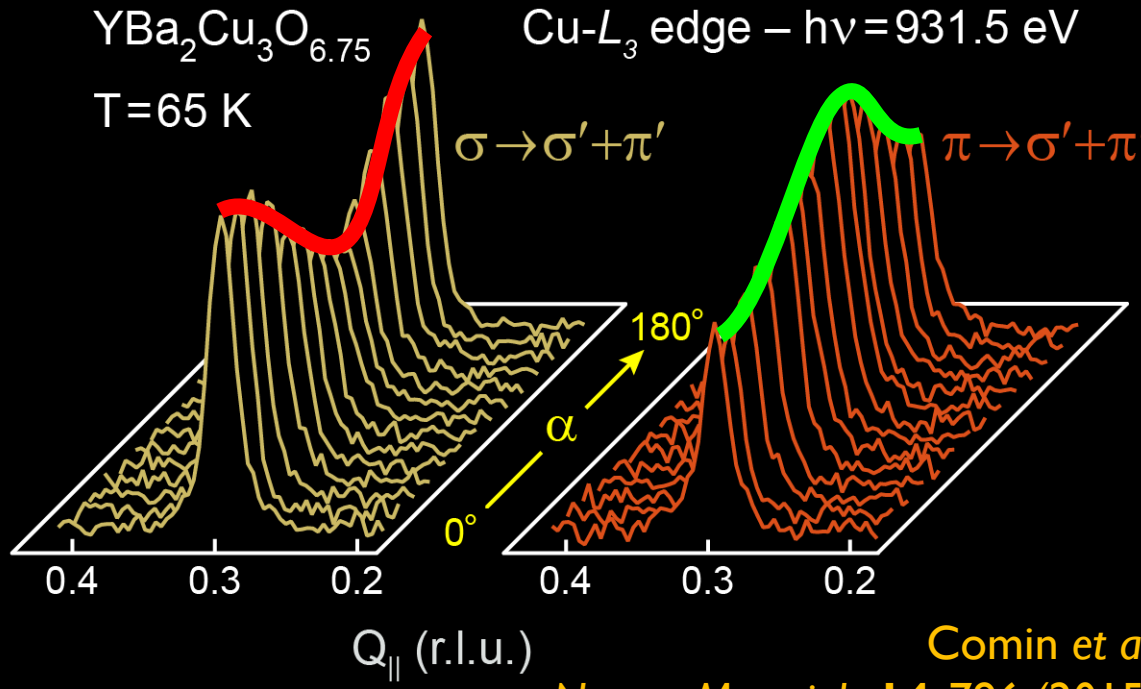
$$I^{\mathrm{RXS}}(\mathbf{Q}, \omega) \propto \left| \sum_{pq} \epsilon_p \cdot F_{pq} \cdot \epsilon'_q \right|^2$$

Revealing the symmetry of charge order in YBCO



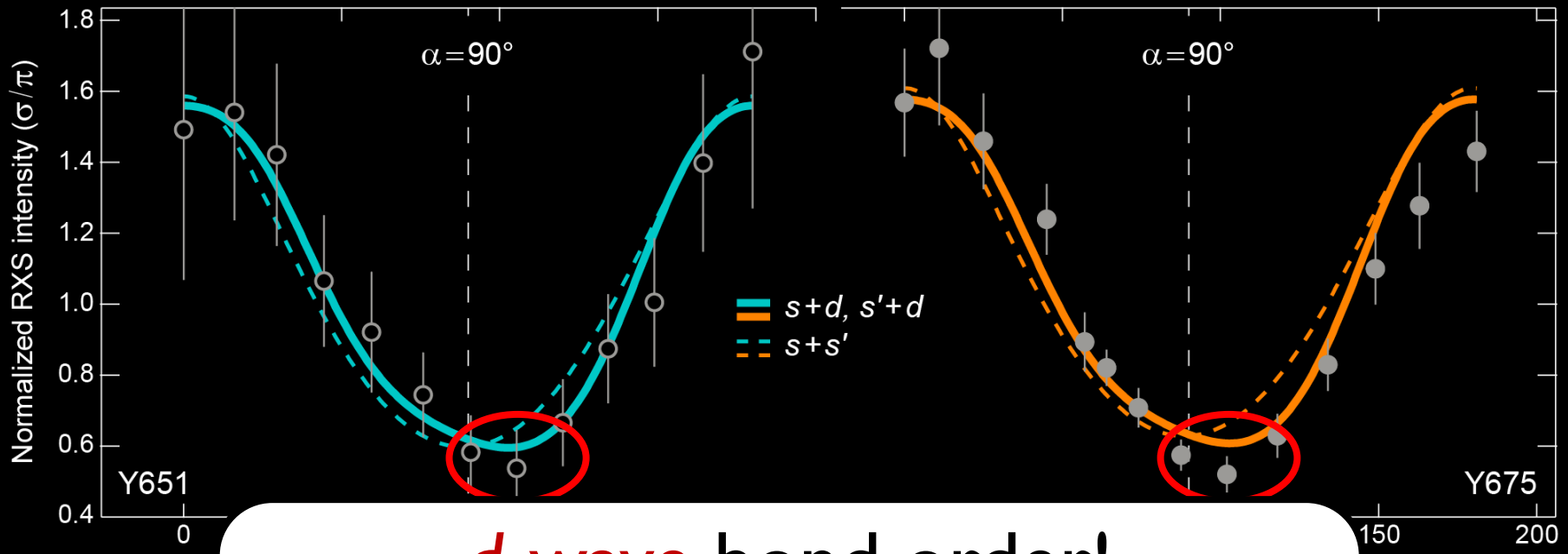


CDW peak

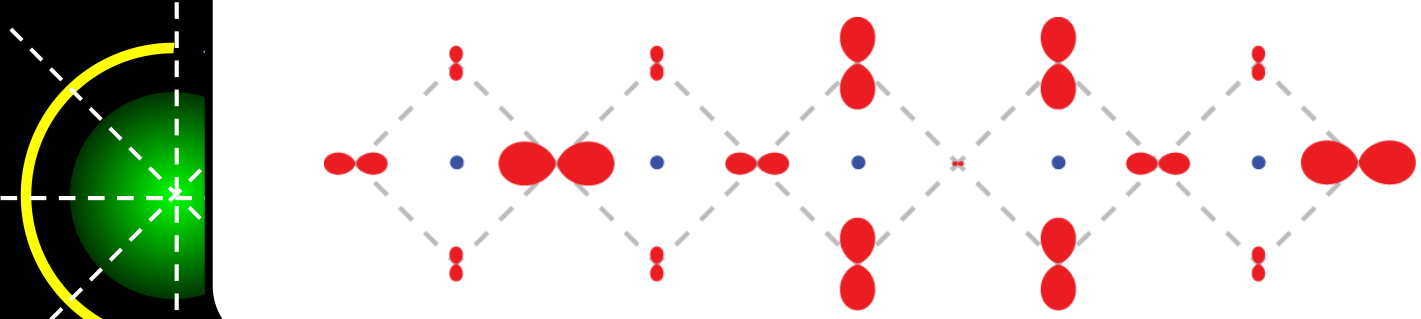


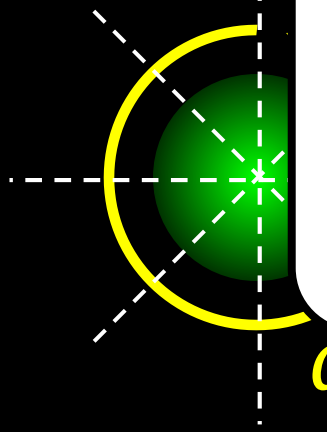
Comin *et al*, *Nature Materials* **14**, 796 (2015)

Revealing the symmetry of charge order in YBCO



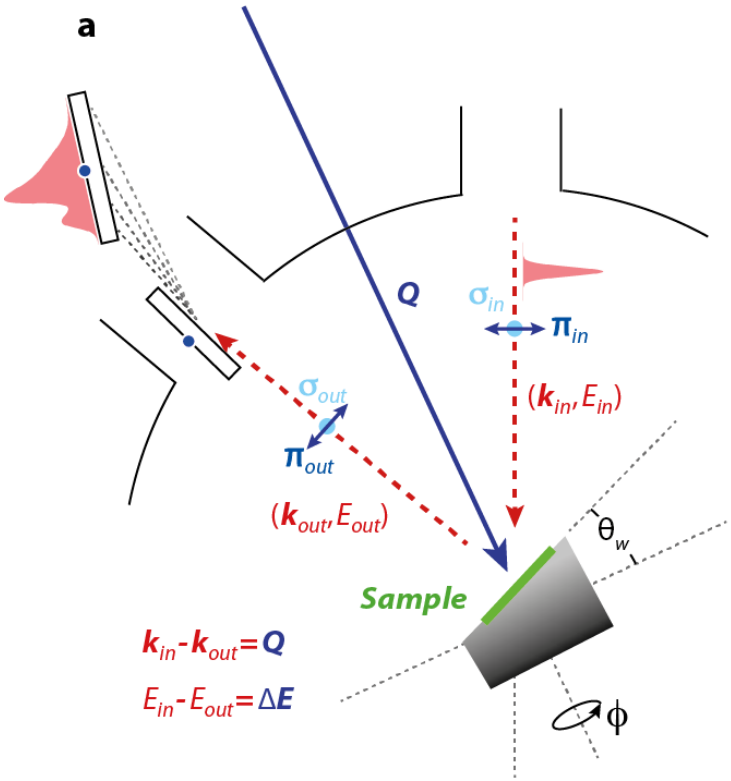
d-wave bond order!

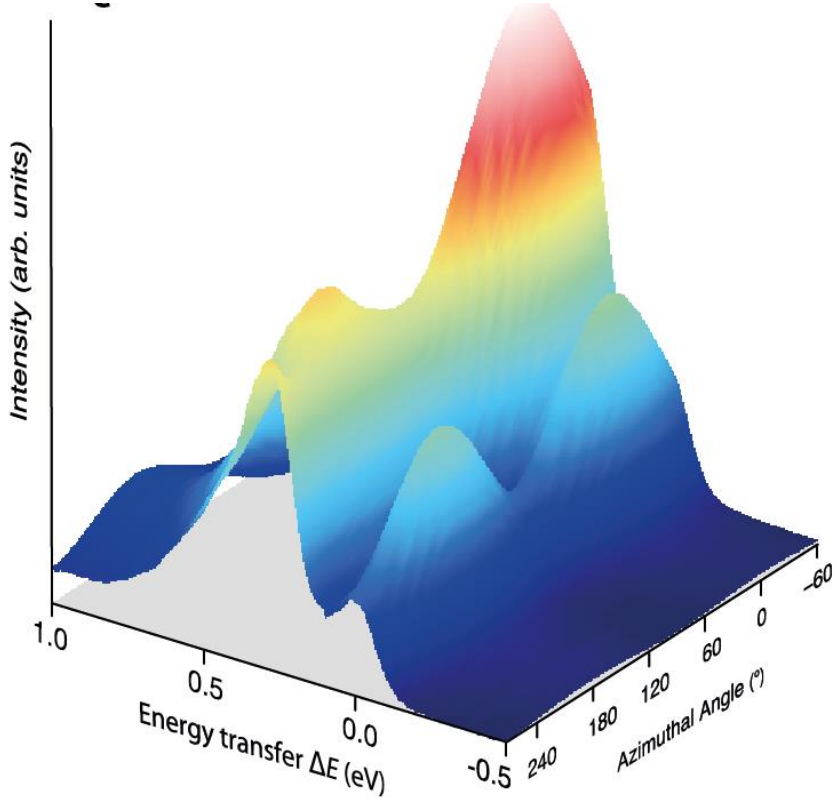




Comin *et al*,
Nature Materials **14**, 796 (2015)

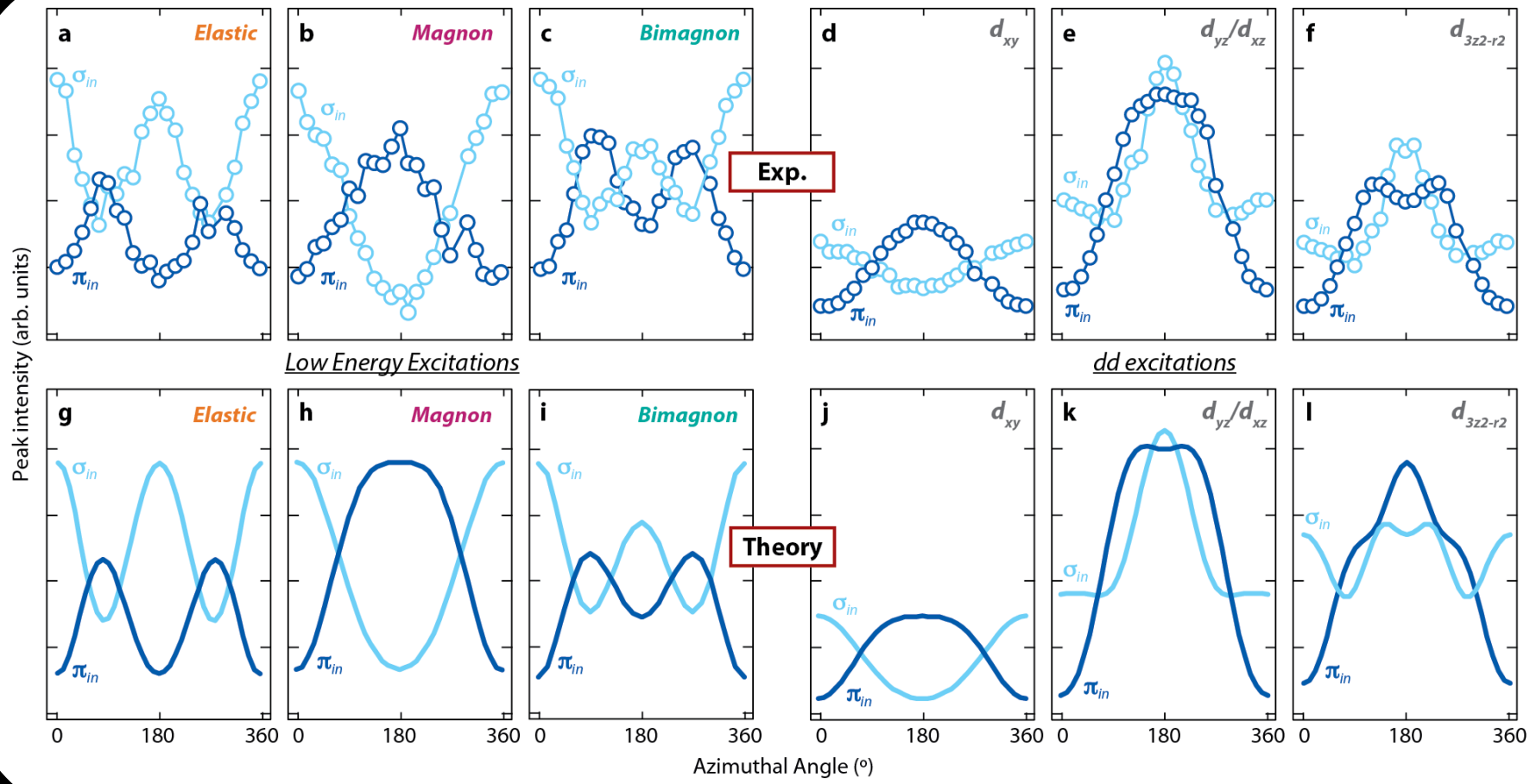
The symmetry of electronic excitations (RIXS)





Kang *et al*,
Under review (2017)

The symmetry of electronic excitations (RIXS)

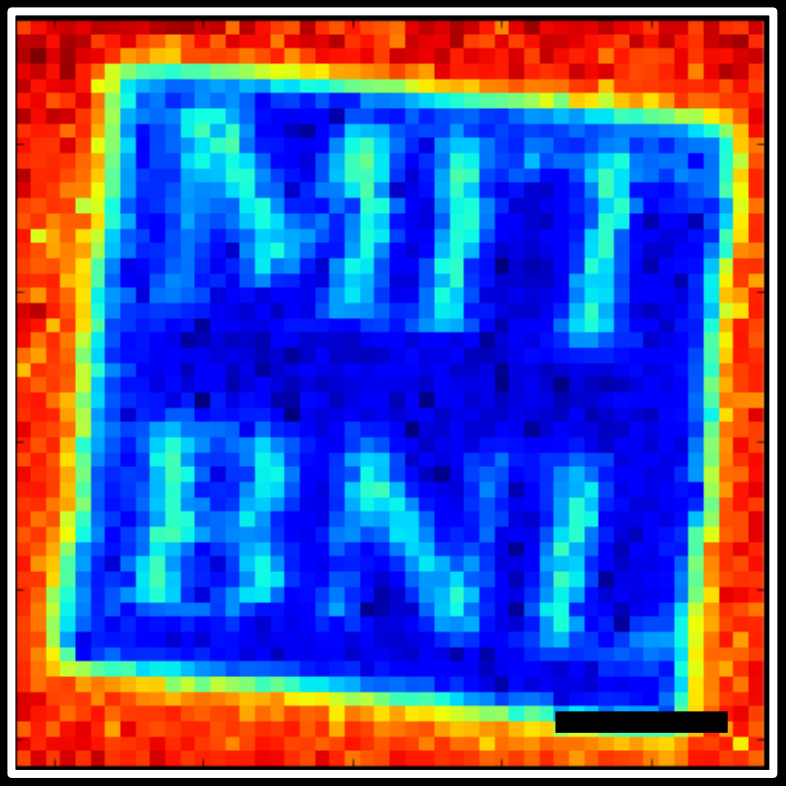


Kang *et al*,
Under review (2017)

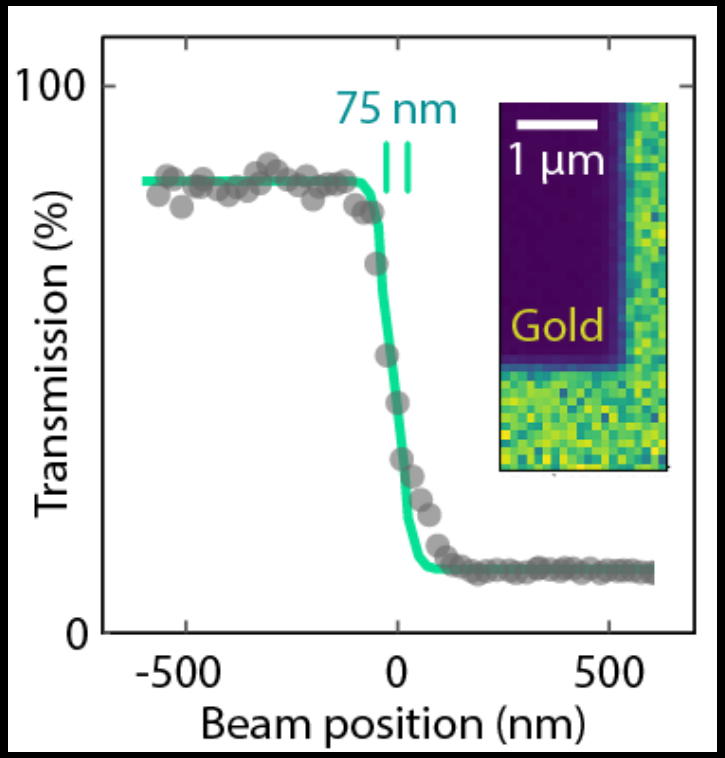
The next frontier:
Coherent scattering

Coherent (nano)scattering

Zone-plate focusing optics: 70 nm spot size



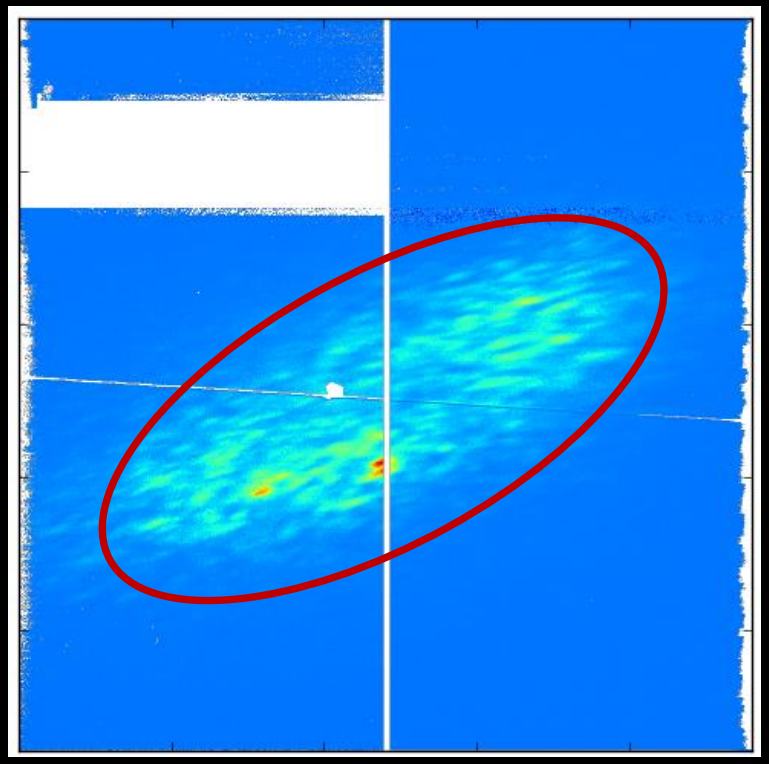
1 µm



Coherent (nano)scattering

First case study: spin-density-wave in $NdNiO_3$ thin films

RECIPROCAL SPACE



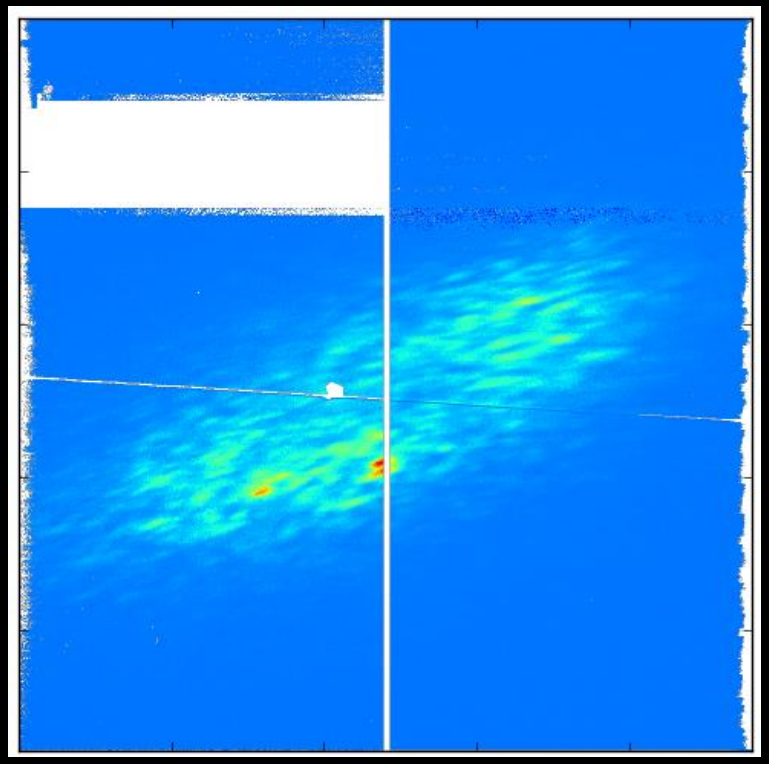
Coherent magnetic scattering from spin-density wave in $NdNiO_3$

Speckle pattern: coherent interference between magnetic domains

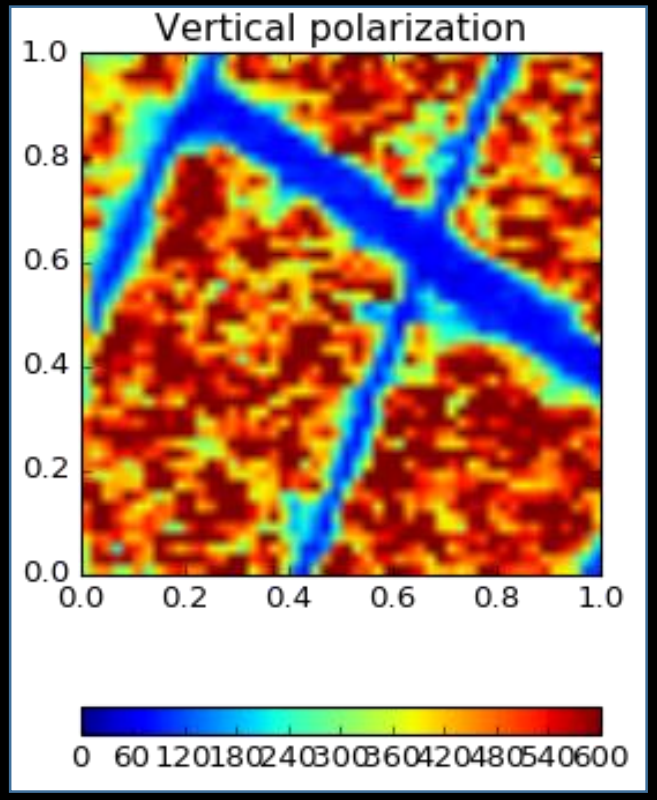
Coherent (nano)scattering

First case study: spin-density-wave in $NdNiO_3$ thin films

RECIPROCAL SPACE ⟶ REAL SPACE (mapping)



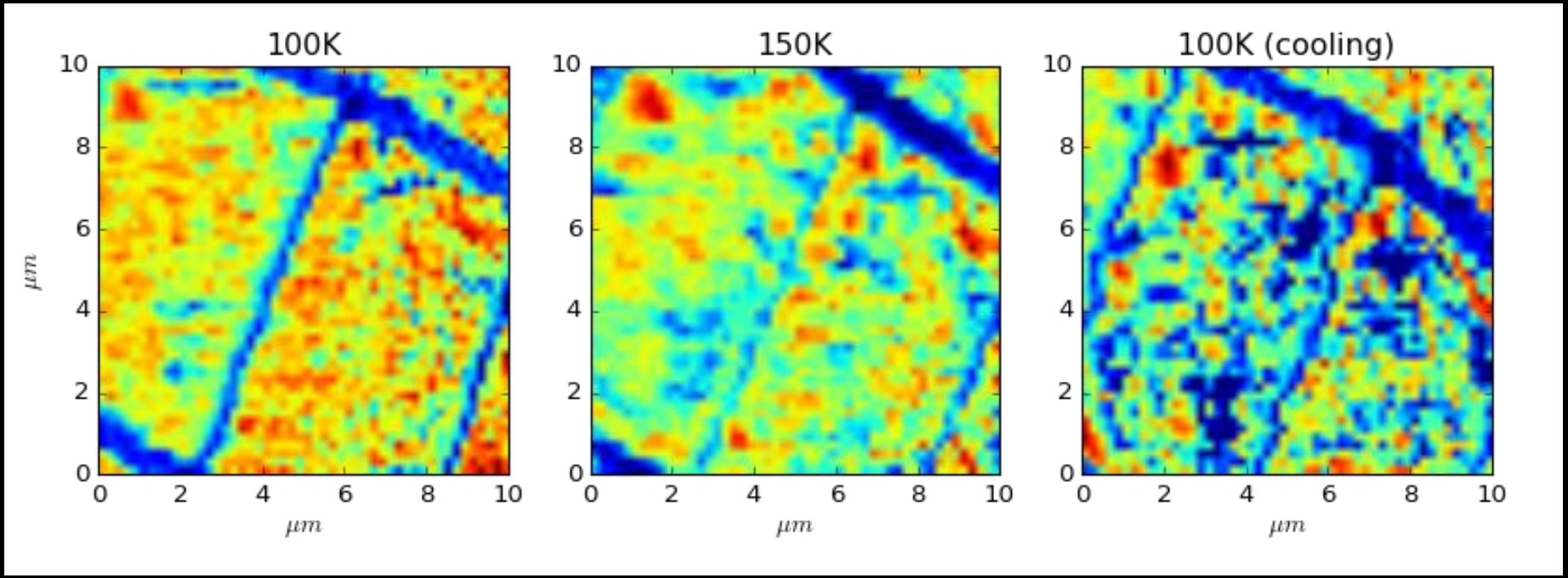
Coherent magnetic scattering from spin-density wave in $NdNiO_3$



Nano-mapping of order parameter

Coherent (nano)scattering

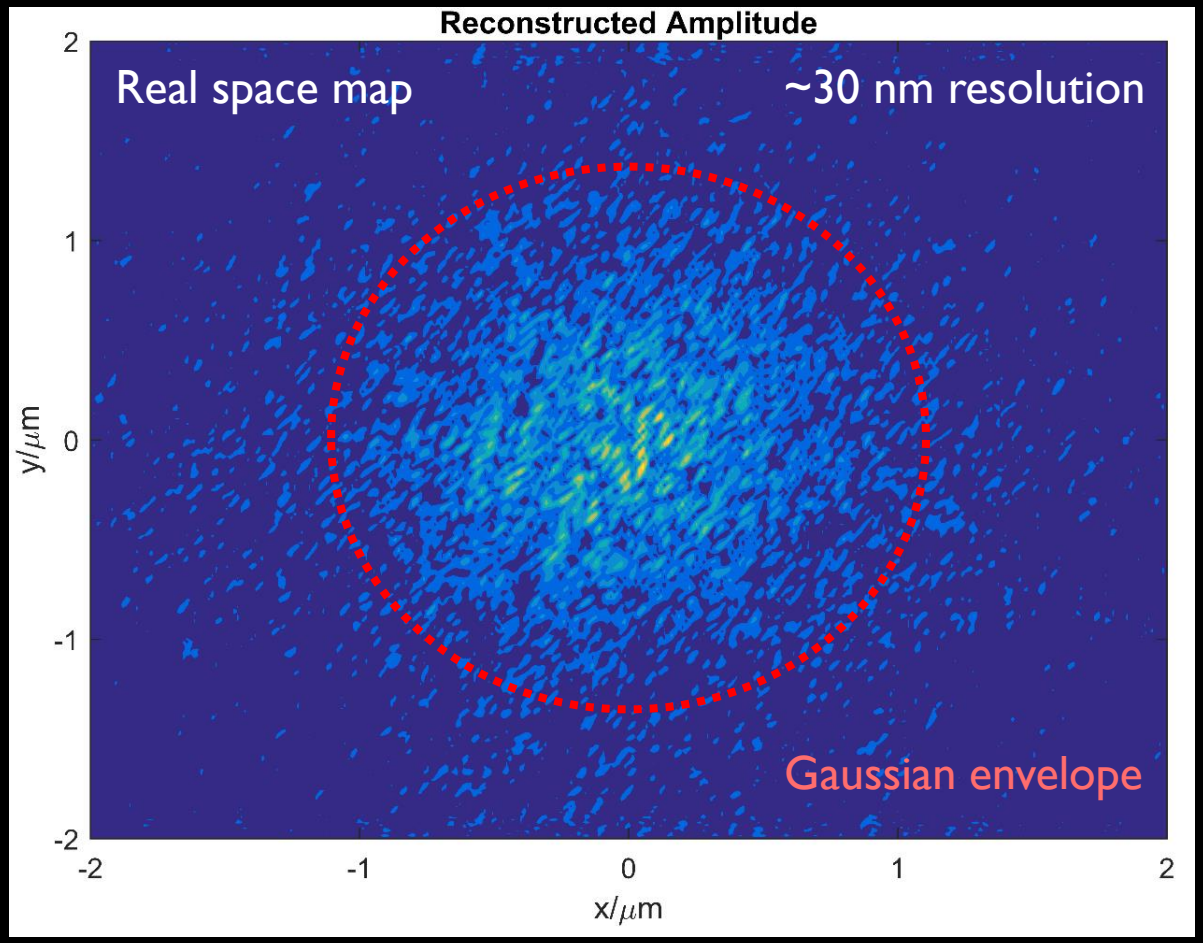
First case study: spin-density-wave in $NdNiO_3$ thin films



Coherent (nano)scattering

COHERENT DIFFRACTIVE IMAGING

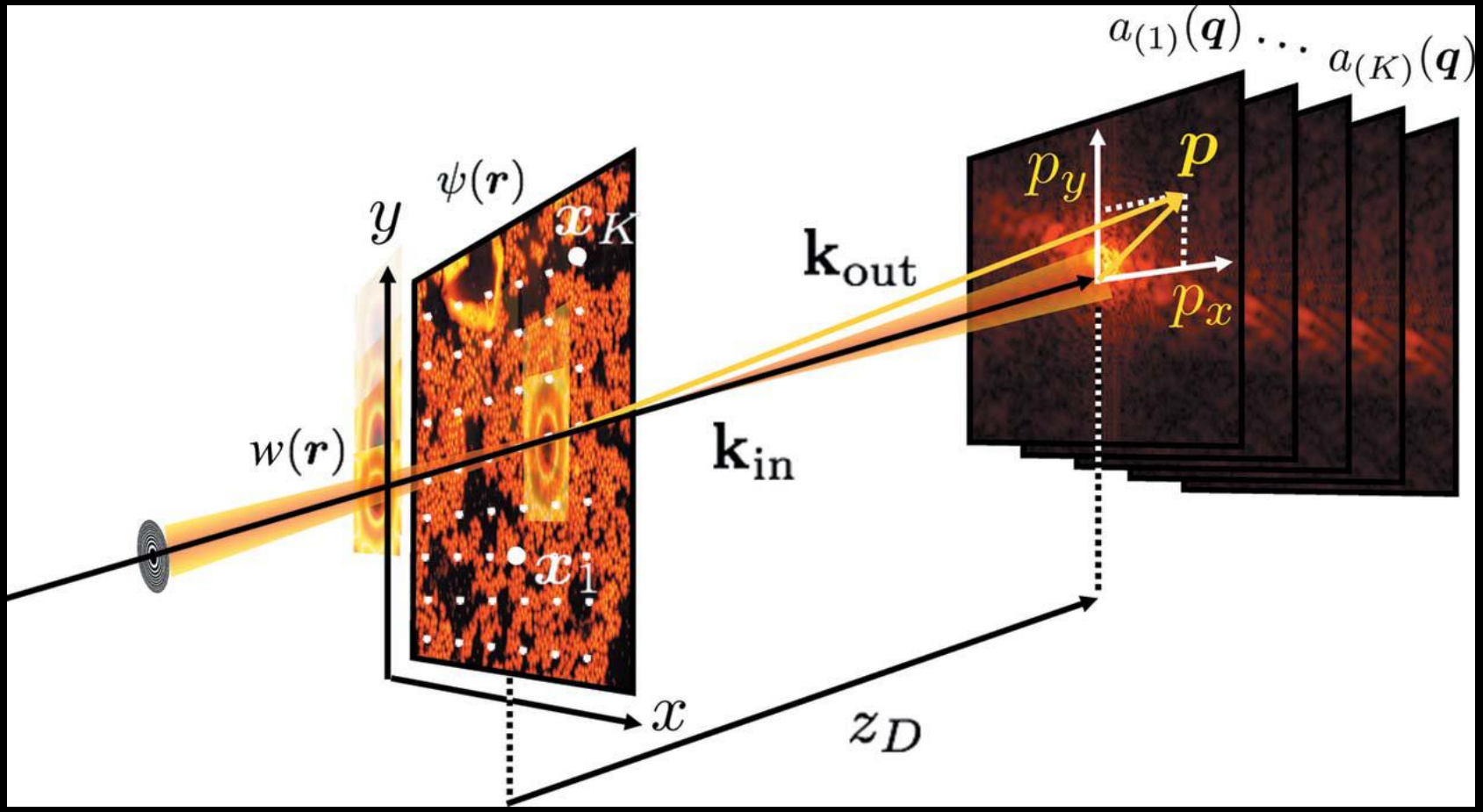
(phase is guessed and optimized to satisfy external constraints)



Coherent (nano)scattering

PTYCHOGRAPHY

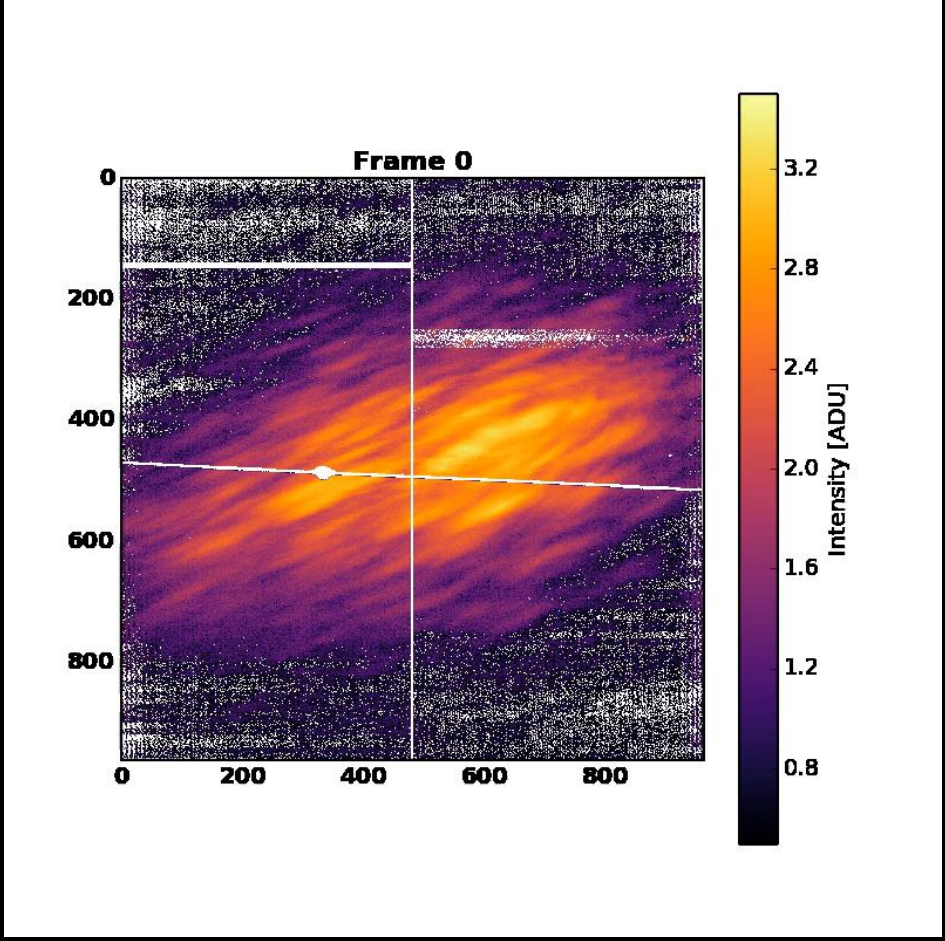
(Scan grid + overlap constraints to retrieve phase)



Coherent (nano)scattering

PTYCHOGRAPHY

(Scan grid + overlap constraints to retrieve phase)



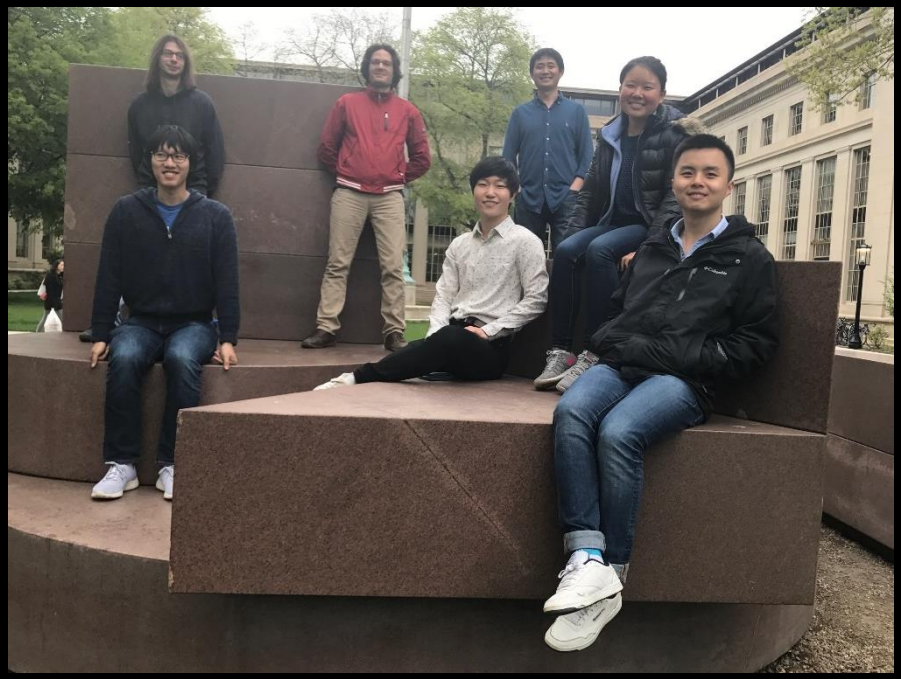
Analysis in progress

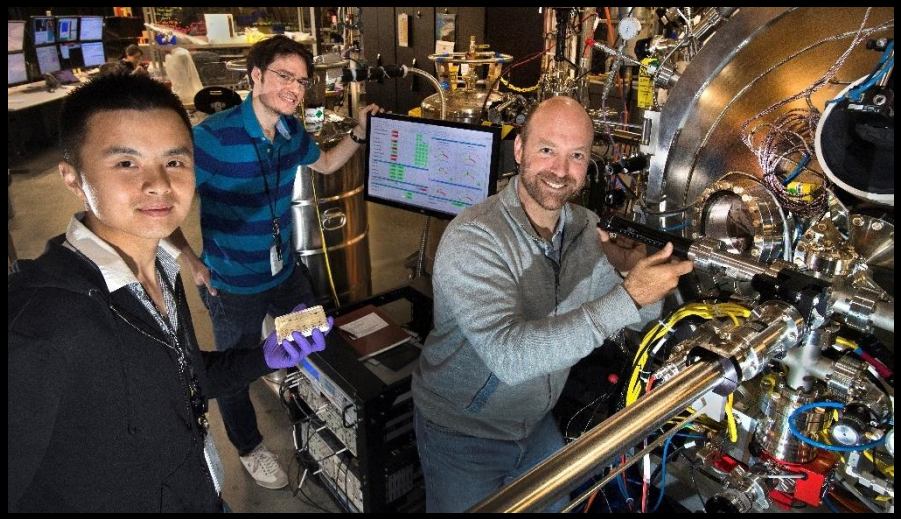
Coherent (nano)scattering

- Highly-coherent soft X-ray scattering provides a very timely opportunity to push the envelope on spatial resolution.

- These new capabilities will prove essential to study naturally inhomogeneous or artificially nanostructured systems.

Acknowledgments





MIT Photon Scattering Group

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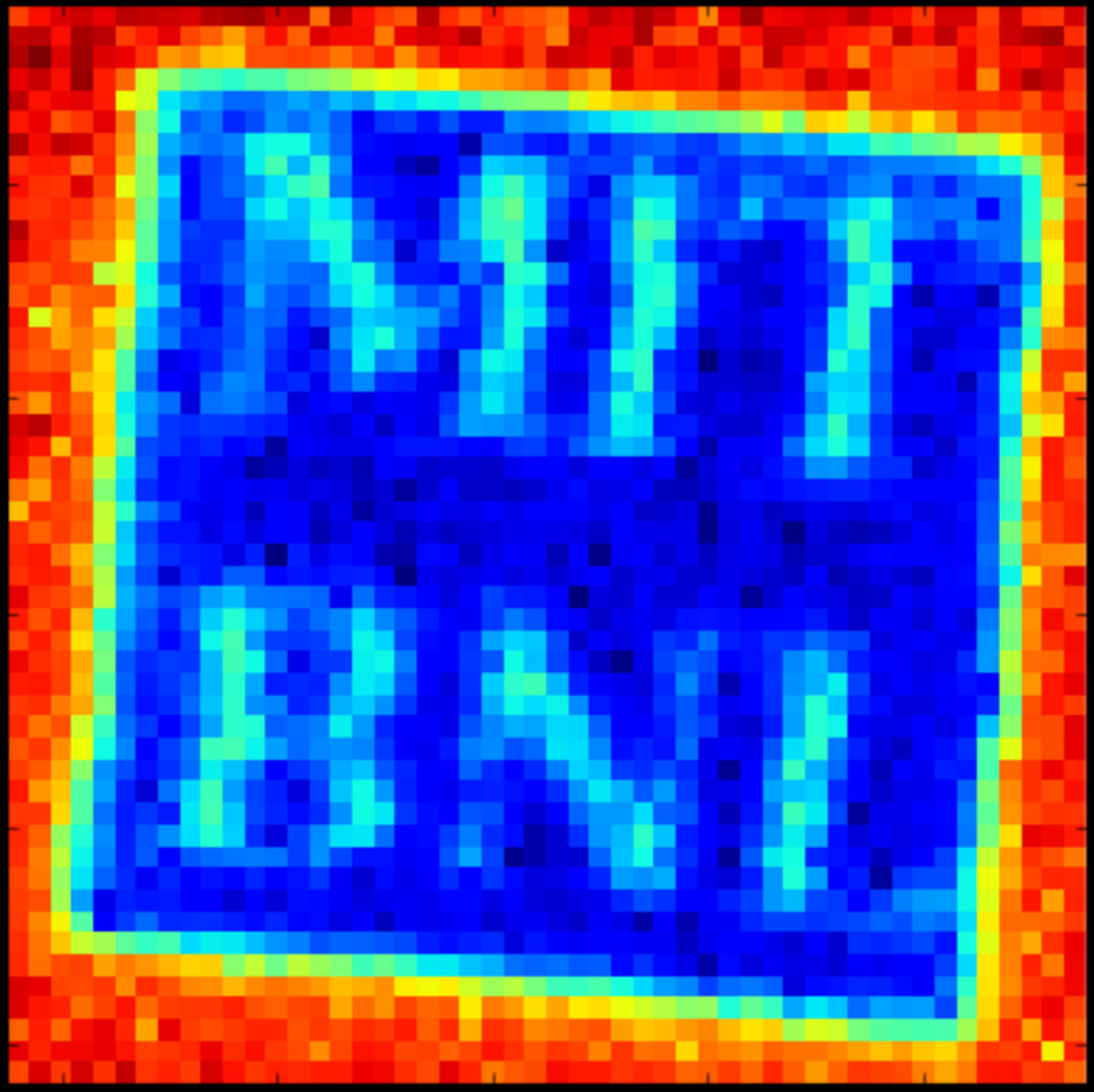
Zhihai Zhu

BNL – beamline CSX-1

C. Mazzoli
A. Barbour
S. Wilkins

Thank you!